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RESEARCH REACTORS AND ALTERNATIVE DEVICES FOR RESEARCH

REPORT OF A CONSULTANTS' MEETING
ON ALTERNATIVES TO RESEARCH REACTORS AS A RESEARCH TOOL
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN
UPPSALA, SWEDEN, 18–21 JUNE 1984

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FOREWORD

The research reactor is a versatile tool, useful in a large number of scientific disciplines and technologies. Over 300 of them are operating world-wide ranging in power from less than one watt critical assemblies to over 100 MW high flux reactors. Most of these facilities particularly in developing countries, are multipurpose reactors used for radioisotope production, neutron activation analysis, training, material science, applied and basic research using neutron beams and other specialized purposes.

A number of countries are considering the purchase of a single multipurpose reactor in the 1-2 MW range or with a thermal flux in the range of $10^{13} ncm^{-2}s^{-1}$. A research reactor of this class is estimated to cost between 4-10 million dollars and much more for higher flux reactors. Because of this high cost and the common nonavailability of adequate resources, these countries may wish to consider alternatives to the reactor, the choice of which would depend on their primary motivation for wanting to acquire a research reactor.

In order to assist these countries in their assessment of the alternatives to research reactors, a Consultants' Meeting was convened by the IAEA to discuss and develop information on the alternatives addressing the principal uses of research reactors including the important function of contribution to the development of an overall infrastructure in basic sciences and technology.

During the period 18-21 June 1984, seven experts from six Member States and staff from the University of Uppsala and the Agency met at the Tandem Accelerator Laboratory, Uppsala, to discuss the above subject. This report includes papers on research reactors, radioisotopic sources, cyclotrons, D-T sources, small accelerators and, in addition, for institutions with limited funds but committed to start a research program, an example of other devices, one of which is described in the paper on 252-Cf plasma Desorption Mass Spectrometry. Spallation neutron sources were specifically excluded due to their advanced technology and high cost of construction.

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AN OVERVIEW

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The topic of this Technical Committee meeting is: Are there any alternatives to research reactors? This question has arisen because there have been a large variety of research reactors of different types and flux levels built during the past three decades. The question is relevant with respect to developing countries who are now proposing to start nuclear research. A research reactor often symbolizes the advent of nuclear research, thanks to the pioneering effort of Fermi and others in 1942. Therefore, any country which builds a research reactor for the first time considers itself as entering the new era of nuclear research and thus nuclear power. However, it may be argued that building a research reactor today is not considered a very involved exercise in technology. Perhaps building an accelerator needs higher technology than building a research reactor. But the fact remains that the research reactor is a sufficiently complex facility which serves diverse purposes especially in a developing country which is desirous of starting a research programme. The process of building a research reactor involves new technologies. In the long run, it is an admirable tool which helps to develop manpower oriented to a nuclear power programme in addition to its uses in Isotope production and as a source of neutrons for research. All these aspects have been discussed before especially in a couple of Technical Committee meetings organized by the Agency last year at Lisbon.

The question that is often asked is whether a research reactor is the ideal facility for basic research in nuclear science. Here one must consider the question of economics, management problems, hazards, etc. There is a feeling that a research reactor could be an unnecessarily expensive, risky tool in the hands of untrained people. Experience during the last couple of decades indicates that this is not so. Many research reactors in developing countries have operated satisfactorily even though their use for research has varied according to priorities of the user.

It is superfluous for me to deal with the details of how a research reactor acts as a source of neutrons for research in both basic neutron physics and other applied areas. Many papers presented in the Lisbon meeting covered these areas. I must, however, emphasize that neutrons from research reactors will continue to be an essential basic tool for new kinds of experiments, exciting the imagination of scientists. Even the smallest research reactor can be utilized for basic fundamental research. An apt illustration of this is what has happened in the 250 KW Austrian reactor being used by Prof. Rauch.

In India we have used the 40 MW research reactor CIRUS providing a flux of 3 x 10^{13}n/cm^2/sec. for the last two decades for basic and applied research. A description of the typical experiments carried out in this facility is given in the appendix. We are now in the process of commissioning a 100 MW D_2O moderated research reactor DHRUVA which will provide neutron flux in the range of 10^{14}n/cm^2/sec. Some of the facilities being built for operation with this reactor are discussed here.

I shall now describe some of the newer types of research reactors we have built. This relates to the reactor called Purnima-2, which is a solution reactor entirely based on U-233. The primary objective of this experiment is to build a reactor with minimum critical mass of fissionable material. This reactor consists of a vessel of zircalloy which contains a solution of uranium nitrate reflected by beryllium oxide and controlled by cadmium control rods. The safety system consists of safety blades and reflector drop mechanisms. The use of highly alpha active U-233 in solution and the system to pump and drain the solution in the glove boxes makes it an interesting but complicated design.

As a follow-up of this reactor, we have designed a reactor called KAMINI (Kalpakkam Mini Pool Reactor). This will again use U-233 alloyed with aluminum in plate type fuel elements. It is designed to operate in the hot cells of the Radiometallurgy Division where fast reactor fuel elements will be examined by neutron radiography. Therefore, this reactor is housed within the heavy shielding in hot cells.

The cost of the KAMINI reactor, apart from the cost of fuel, is only Rs. 60 lakhs or equivalent to 600,000 dollars. It is entirely designed and built in India and can operate at a power level of 50 - 100 KW. This can be used for neutron radiography as well as for activation analysis. Some experiments on dosimetry are also planned. The reactor will therefore serve as a useful facility for applied nuclear research programmes of a Centre devoted to development of fast reactor technology.

Many alternatives to a research reactor as a neutron source will be discussed at the meeting. Californium-252 as a neutron source has been in use for several years now. However, it is the cost of making this source and the limited flux of neutrons that sets limitations in its use for a variety of purposes. Pulsed neutrons using accelerators have been used very effectively, especially in nuclear physics research.

Intense pulses of neutrons can also be obtained from other sources like plasma focus. Here the stored energy in a condenser bank is discharged through a plasma tube ideally designed to produce short bursts of neutrons by D-D or D-T reactions. While detailed scientific understanding of the process leading to neutron production is still lacking, many groups have built such sources and upgraded them to reach neutron intensities of the order of 10^{13}n/pulse. Further technical advances may increase the repetition rate. This may then turn out to be an alternative source of low-cost neutrons for scientific research.

There are other important methods to improve the intensity of neutrons. These are based on high energy accelerators in which the spallation reaction is made use of. The SNS facility in the Rutherford Laboratories will be the first of its kind producing a very intense source of neutrons in the near future. However, I am given to understand that the competitive attempts in Los Alamos, using a proton storage ring, may reach even higher intensities. Thus there are a great variety of facilities which are becoming plausible alternatives to research reactors.

Accelerators and fusion devices seem more complicated compared to the research reactor. A low power research reactor is simpler and could be operated and used by a small number of scientific and technical people in a developing country. The experimental research reactor will continue to be useful in the next couple of decades until other technologies in fusion produce elegant alternative neutron sources. The need to train manpower through a research reactor for any country planning to introduce nuclear power reactors will prove to be an important reason in opting for the Research Reactors.

CHARACTERISTICS AND USES OF A 250 kW TRIGA REACTOR

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Abstract

The 250 kW TRIGA Mark II reactor is a light water reactor with solid fuel elements in which the zirconium hydride moderator is homogeneously distributed between enriched uranium. Therefore the reactor has the large prompt negative temperature coefficient of reactivity, the fuel also has very high retention of radioactive fission products. The reactor core is a cylindrical configuration with an annular graphite reflector. The experimental facilities include a rotary specimen rack, a central in-core radiation thimble, a pneumatic transfer system, and pulsing capability. Other experimental facilities include two radial and two tangential beam tubes, a graphite thermal column, and a graphite thermalizing column. At the stady state power of 250 kW the peak flux is 1×10^{13} n/cm²s in the central test position. In addition, pulsing to about 2000 MW is usually provided giving peak fluxes of about 2×10^{16} n/cm²sec. All TRIGA reactors produce a core-average thermal neutron flux of about 10^{7} n.v per watt. Only with very large accelerators such a high neutron fluxe could be achieved.

In order to give an appreciation for the research conducted at research reactors, the types of research could be summarized as follows: thermal neutron scattering, neutron radiography, neutron and nuclear physics, activation analysis, radiochemistry, biology and medicine, and teaching and training. Typical applied research with a 250 kW reactor has been conducted in medicine in biology, archeology, metallurgy and materials science, engineering and criminology. It is well known that research reactors have been used routinely to produce isotopes for industry and medicine. In some instances, reactors are the preferred method of isotopes production.

We can concluded that the 250 kW TRIGA research reactor is a useful and wide ranging sources of radiation for basic and applied research. The operation cost for this instrument is relatively low.

1. Introduction

During the early years of nuclear development, many low or medium flux research reactors were set up in countries throughout the world. These played a vital role in the establishment and development of nuclear technology, as well as contributing importantly to research in other fields. Nevertheless, the following question is usually asked: "Should we operate a research reactor?" We believe that the answer must be positive. Every country should operate a research reactor or at least participate in the performance of scientific research activities using one type or another of research reactors. The reason behind is the fact that nuclear energy and its applications have proven to possess great capabilities in contributing to the well being of mankind and seems to still have greater potentials for the near and far future. The exploration of these potentials is not the responsibility of only a group of countries. This responsibility must be shared by all nations, each within its own best technical and economical capabilities. To do so, all means and tools must be utilized, including research reactors. To-day the research reactor as used in many laboratories and research centres is a useful and wide ranging source of radiation for a scientific research and for applications. Thermal neutron reactors in the power range from less than 10 kW to more than 100 MW have been used.

The most widely used research and test reactor in the world is the TRIGA reactor. Sixty-three TRIGA reactors have been or are being built on five continents. The massed output of research includes results to be found in all aspects of modern life, including food preservation, production of food bearing plants, nuclear medicine, industrial quality control through nuclear measurements, training of engineers and power reactor operators, and direct use of neutrons in research such as solid state physics, chemistry, physics, biology, medicine and neutron radiography. It should be pointed out that the research`s intellignece and ingenuity together with even a relatively small reactor system can produce a wide variety of significant experimental results.

2. TRIGA Research Reactor

Development of the TRIGA reactors began at General Atomic in 1956. It was recognized that there was a need for a research reactor which provided inherent safety, flixibility of operation and economy for research institute

and university application. These became the underlying objectives in the development work. In addition to achieving these objectives, it was necessary to also provide the user with appropriate experimental facilities and a high useable neutron flux.

The development work centered around the use of a uranium-zirconium-hydride (UZrH) fuel which had the potential for meeting all of the objectives. Most importantly it would provide a reactor with a large prompt negative temperature coefficient for inherent safety against reactivity accidents. It also had very high retention of radioactive fission products limiting the potential for radioactive release into the reactor environment.

The development of the UZrH fuel for this application was achieved and a critical experiment was operated in 1957 which provided proof of operating characteristics for the TRIGA reactors. The first reactor went into operation at General Atomic in 1958 at a power level of 10 kW.

Further experiments proved that the large prompt negative temperature coeficient of the reactor made it well-suited to "pulsed" operation as well as steady-state. Originally routine pulsing was to about 2000 times the steady-state power. The development of the TRIGA reactor design and the fuel capability has continued since 1958 to provide reactors with enhanced and diverse capabiliteis, including thermal power levels up to 50 MW steady-state and the ability to pulse to peak powers of over 20,000 MW.

The unique features of the TRIGA reactor have resulted in it being the most widely used research and test reactor in the world with 63 reactors being sold in 20 countries around the world. Eleven of these TRIGA reactors are conversions of reactors which originally used MTR plate-type fuel. This provided the owners with the enhanced safety and operational characteristics as well as the ability for pulsed operation.

All of the TRIGA reactors are open pool, light-water-cooled reactors using UZrH fuel. The original reactors put into operation at General Atomic and at the University of Arizona in 1958 have a configuration in which the reactor is placed at the bottom of the open cylindrical tank with the top of the tank at ground level. Access to the core is from the top of the pool only. This configuration, designated as the Mark I, is still sold today and is the simplest and the least expensive model available.

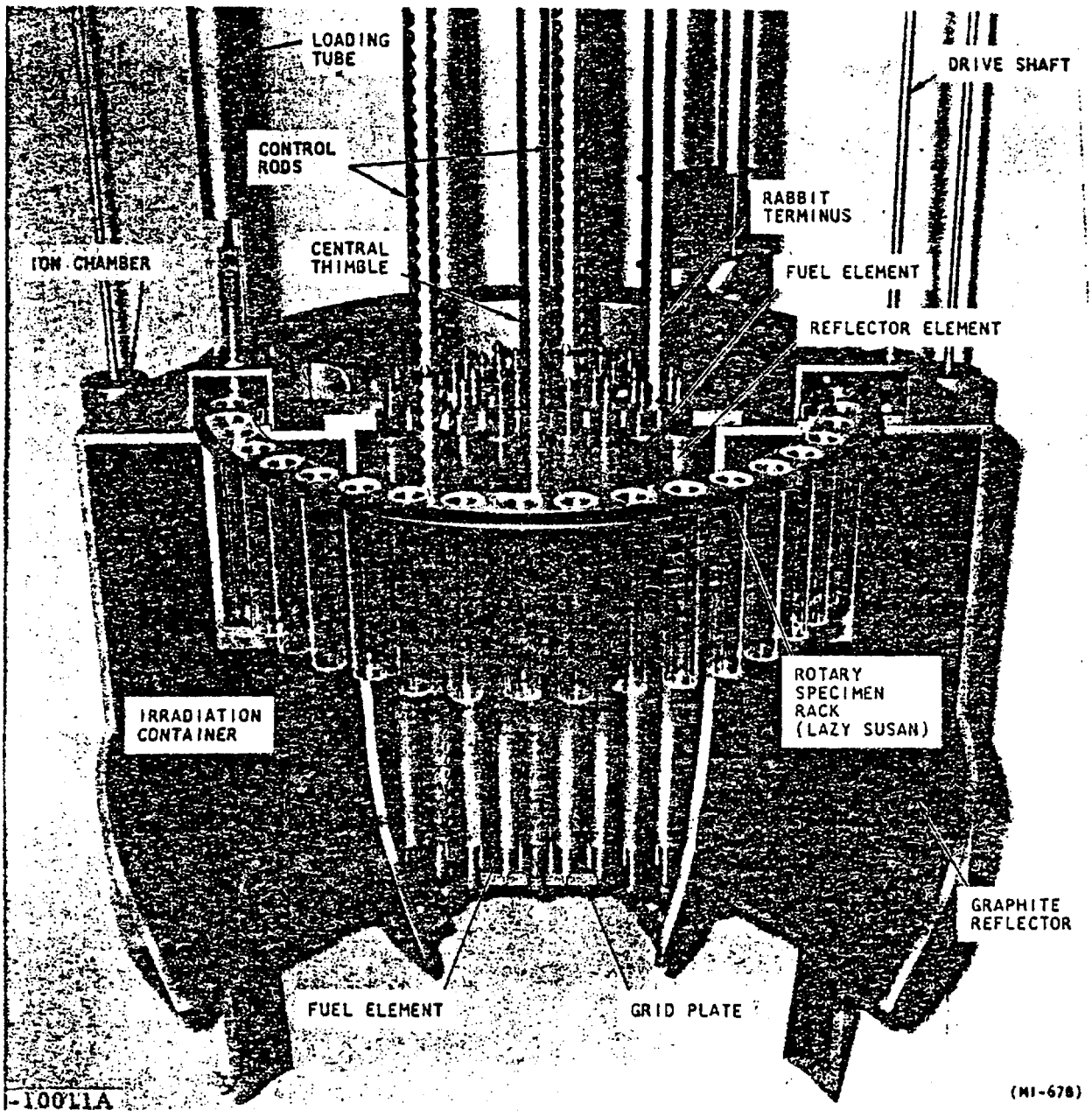

Fig. 1. TRIGA Mark I and Core Configuration

The configuration of the Mark I is shown in Fig. 1. The reactor core is a cylindrical configuration with an annular graphite reflector. The experimental facilities generally include a rotary specimen rack, a central in-core irradiation thimble, a pneumatic transfer system, and the ability for inserting experiments in various in-core locations, and pulsing capability. The steady-state power level can be as high as 2000 kW with natural convection cooling and can be increased to 3000 kW with forced downflow cooling. Pulsing to about 2000 MW is ussually provided.

An additional level of experimental capability was added by placing this same basic reactor within a shield structure above the floor level, allowing horizontal access to the core. This allowed incorporation of radial and tangential beamports as well as a graphite thermal column. The reactor design and the control and instrumentation system for Mark I and Mark II are otherwise nearly identical.

The Mark II configuration is the most widely used TRIGA with 20 currently in operation. The first Mark II reactors started operation in 1960 in Rome, Italy and at the University of Illinois. A 3000 kW Mark II is currently under construction.

The ultimate in experimental flexibility in a low-powered research reactor was achieved by the Mark III design. In addition to extensive beamport facilities, the Mark III incorporates a large exposure room and a large reactor pool with a movable reactor core (Fig. 2). The reactoris supported from a movable bridge, is water reflected, and operates at power levels up to 2000 kW with natural convection cooling. The first Mark III prototype was put into operation at General Atomic in 1965 and the first Mark III built commercially went critical in 1968 at the University of California, Berkeley.

The most recent addition to the TRIGA family is a 50 MW multipurpose test reactor which provides high fluxes for power reactor fuel testing, isotope production and beamport experiments. This reactor provides a very versatile facility retaining the safety features of the UZrH fuel, provides long core life with low enriched fuel, and does it all very economically.

A dual core facility for power reactor fuel development testing has been put into operation in Romania. This facility contains a 14 MW steady-state reactor and an ACPR within the same pool. The 14 MW core is designed to include three in-core test loops with thermal fluxes in test positions exceeding 10^{14} n/cm² sec.

3. Research and Training with 250 kW TRIGA Reactor

In order to give an appreciation for the research conducted at research reactors, the types of research are summarized here. This research has been heavily centered around, but not limited to, thermal neutron scattering (both elastic and inelastic), neutron and nuclear physics, and small angle

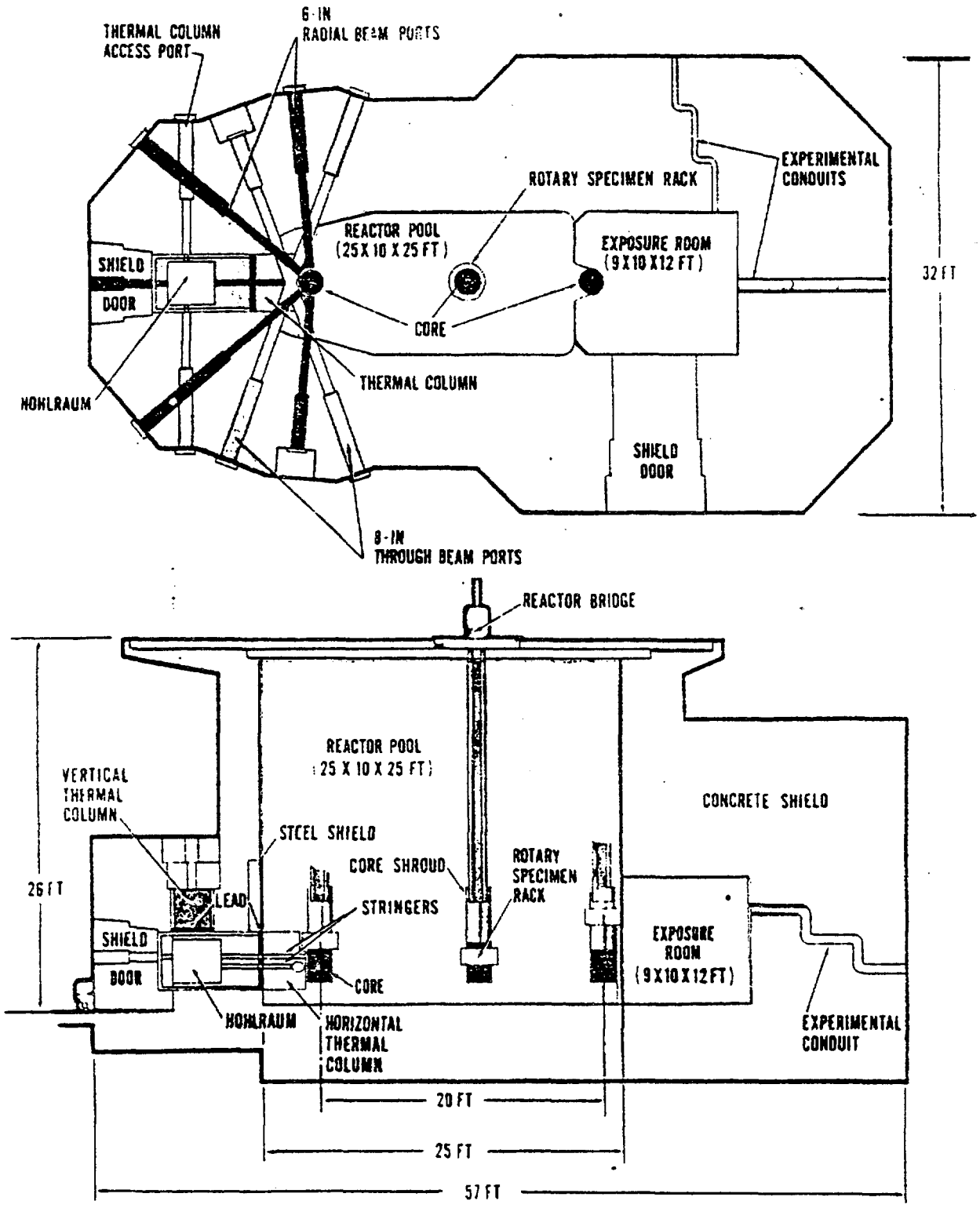

Fig. 2. TRIGA Mark III Configuration

neutron scattering. This research uses the special feature of slow neutrons (with energies from 10^{-7} to 0.5 eV) in which a neutron can have an energy comparable with molecular and crystal excitations and simultaneously a wavelength comparable with interatomic or intermolecular spacing. Extensive work therefore involves the investigation of coherent neutron scattering using predominantly thermal neutrons but also, on occasion, cold neutrons. Neutrons can be selected with relative ease with energies suitable for investigations of very weak forces (10^{-7} eV) to those for investigation of strong binding forces (0.1 - 0.5 eV). In addition to aiding in the conduct of research, the TRIGA research reactor also offers splendid opportunities for the training of personnel for nuclear technology. The inherent safety of the TRIGA reactor aids enormously in increasing the variety of tests can be conducted as part of a manpower training effort. Table 1 lists specialized basis research including: slow neutorn scattering, nuclear and neutron physics, radiochemistry, biology and medicine, and teaching and training. Table 2 lists typical applied research with a TRIGA reactor. Many of the research areas listed as applied research use the basic techniques set forth in Table 1.

TABLE 1

BASIC RESEARCH WITH RESEARCH REACTOR

Slow Neutron Scattering

- Crystal and powder diffraction, including polarization and magnetic studies
- Quasi-elastic scattering
- Inelastic neutron scattering
- Small angle and diffuse scattering

Nuclear and Neutron Physics

- Neutorn interferometry
- Ultra-cold neutron physics
- Nuclear gamma ray studies
- Conversion electrons
- Pair formation

TABLE 1 cont.

Radiochemistry

- Studies of fuel burnup
- Mass spectrometer for fission products
- Recoil atom chemistry
- Isometric states
- Double capture events

Biology and Medicine

- Cell research with filtered, high energy neutron beams
- Cancer research and therapy with 2 KeV neutrons

Teaching and Training

- Demonstration of reactor parameters involving delayed critical experiments
- Teachniques of nuclear research for biologists, chemists, physicists, and engineers

TABLE 2

TYPICAL APPLIED RESEARCH WITH RESEARCH REACTOR

Petroleum

- Analyzing oil refinery feed stock for impurities, e.g., vanadium
- Reactor produced isotopes for tracer studies in performance of catalytic crackers, location of foam in crude oil lines, and in chemical reactors
- Tracer experiments in oil fields (T_2O, Kr^{85})
- Emulsification, demulsification of oil and water

Medicine and Biology

- Analysis of fingernail samples for high concentrations of sodium for early detection of cystic fibrosis in children
- Trace element determination in various biological samples in relation to specified diseases
- Utilization of reactor-produced isotopes (e.g., gold, iodine, technetium) in clinical diagnostic and therapeutic medical applications

TABLE 2 cont.

- Small angle scattering studies of protein structure and conformation
- Treatmetn of brain tumors

Archeology

- Origin and trade routes for coins
- Degrees of debasement of silver and gold coins
- Origin of clays and pottery through trace elements
- Neutron radiography of specimens (nondestructive tests)

Isotope Production

- Medical isotopes (gold, iodine, technetium)
- Isotopes for biological research
- Industrial isotopes

Metallurgy and Materials Science

- Neutron diffraction studies of steel (short-range-order)
- Boron distribution in special steels
- Relation of corrosion to crystal structure in steels and aluminum alloys
- Diffusion in alloys and two-phase systems
- Phosphorus doping of silicon
- Use of reactor-produced istopes in wear studies
- Nuclear fuel studies
- Small angle scattering analysis of extended defects (e.g., precipitate zones, vitreous substances, fatigue, recrystallation, creep, dislocation density, internal voids)
- Texture formation in metals and alloys
- Hydrogen diffusion in metals (quasi-elastis scattering)

Engineering

- Safety tests of nuclear power reactor fuel
- Radiation damage in materials

Cryminology

- Determination of gunpowder residues
- Matching of evidence-type materials (paint, glass, grease, paper, plastic, rubber, hair, tissue, etc.)

A pulsed TRIGA research reactor offers numerous area of research not open to even the largest steady state research reactors. The pulsed reactor excels as a neutron source for a number of experiments when for example:

1. the output signal is proportional to the square of the neutron flux (as in a variety of double events);
2. the effective cross sections are very small or the half lives are very short (isometric states); or
3. fluxes considerably larger than 10^{15} n/cm² sec are required.

All pulsed TRIGA reactors produce instantaneous peak fluxes in core of 10^{16} - 10^{17} n/cm² sec. Table 3 lists a number of applications suited to pulsed reactor utilization.

TABLE 3

PULSED REACTOR UTILIZATION

Radiochemistry

- Studies of short-lived isomeric states
- Studies of double capture events

Physics

- Neutron-neutron scatter experiment ($nv \geq 10^{17}$)
- Neutron bottle experiments - fundamental neutron parameters
- Lasr pumping

Training and Teaching

- Testing to destruction of power reactor nuclear fuel
- Testing of power reactor electronic control circuits under "accident" conditions
- Pulsed neutron radiography for transient phenomenon (e.g., two-phase diffusion or events lasting 1 millisecond).

4. Conclusion

Concerning the TRIGA reactor we can conclude that it is very good experimental installation because the experimental and irradiation facilities are extensive and versatile, physical access and observation of the core are possible at all times through the vertical water shield, and the prompt negative temperature coefficient of reactivity is very large which gives TRIGA its built-in safety. In addition, thelow operational cost of the reactor (about $ 200.000 per year) makes it suitable for use in developing and developed countries for training, research and isotope production. We believe taht no other experimental equipment can replace the research reactor as neutron sources because they are capable of producing very high fluxes of neutorns having a considerable range of energies, from a few meV to 10 MeV, and basic training on a research reactor provides an essential understanding of the nuclear process. The training given to engineers, scientists and technicians should take the form of performing creative work in order to obtain practical experience and technical confidence.

RADIOISOTOPE NEUTRON SOURCES – CHARACTERISTICS AND APPLICATIONS

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Abstract

Neutron sources that employ radioactive materials are characterized by several properties that can lead to their selection for certain applications in neutron physics and industrial applications. These properties include portability, low cost, and a predictable and stable neutron yield when compared with accelerator-based neutron sources. Some fundamental limitations include the need to handle relatively large samples of radioactive materials, a restricted range of available intensity, and a fixed and often broad neutron energy spectrum.

Radioisotope neutron sources fall into three main categories: spontaneous fission sources, photoneutron sources, and neutron sources based on the (alpha,n) reaction in beryllium. The neutron yield, energy spectrum, and special properties of each of these categories are reviewed in this paper.

Spontaneous Neutron Sources

Many heavy nuclides are known to undergo spontaneous fission, emitting fast neutrons in the process. Observational data are available covering atomic numbers ranging from 90 through 107 and half-lives of the decay from less than 1 second through 10^{17} years. The number of

neutrons emitted per fission event varies considerably from a low of about 1.3 to as high as 4 for some transuranic elements. An evaluation of these physical properties together with potential availability of the isotope has led to the selection of 7 nuclides as primary candidates to serve as practical neutron sources. These are ^{240}Pu, ^{242}Pu, ^{244}Cm, ^{246}Cm, ^{248}Cm, ^{252}Cf and ^{254}Cf.

Of these candidates, the isotope ^{252}Cf has achieved by far the greatest popularity. This stems from its good combination of half-life and neutron yield per unit mass, and its reasonable availability through production in high flux reactors. After chemical separation, the californium is fabricated into practical neutron sources through a number of different procedures. One method involves electroplating the material from a dilute nitrate solution onto a platinum-iridium wire. Alternatively, the californium can be coprecipitated with iron as a hydroxide, converted to an oxide by heating, and compressed into a small pellet. This active material is then carefully encapsulated, generally using platinum, stainless steel, aluminum, or Zircaloy. Because only micrograms of californium are sufficient to make sources of useful intensity, the encapsulation often dominates the physical mass of the finished source.

Some important properties of ^{252}Cf are summarized in Table 1. Only about 3% of the ^{252}Cf decays are by spontaneous fission. The remainder proceed by alpha emission. In encapsulated sources, the wall thickness is sufficient to stop both the fission fragments and the

TABLE 1 Properties of Californium-252

Neutron yield	2.34×10^{12} n/s per gram 0.116 n/s per Bq (total activity)
Neutrons per fission	3.75
Average neutron energy	2.14 MeV, fission spectrum
Half-life	2.65 years
Effective half-life spontaneous fission only	85.5 years
Heat generation	39 W/g

alpha particles from these decays, leaving only the fission neutrons and gamma rays to emerge from the source. Sealed californium sources are available that contain between a few micrograms to 5 milligrams of ^{252}Cf. Maximum neutron yield from a single source is therefore about 10^{10} neutrons per second.

The neutrons produced from a californium source are distributed in energy over a typical fission spectrum. Because the sources are usually physically small, little scattering or other neutron interactions take place within the source itself, and the emerging neutrons have an energy distribution essentially the same as upon emission in the fission process. The model most often used to describe this distribution is the Maxwellian:

$$N(E) = Const * E^{1/2} \exp(-E/T)$$

Experimental measurements of the neutron spectrum generally yield an average neutron energy of about 2.12 MeV. Most measurements tend to confirm the generaly

Maxwellian shape for the spectrum, although measurements are difficult at the low and high energy extremes.

The californium used to fabricate neutron sources is seldom isotopically separated. Hence one can expect a mixture of californium isotopes with a relative abundance depending on the method of production. The fraction of ^{252}Cf can range anywhere from between about 45% and 85%.(Ref. 1) Significant amounts of californium isotope numbers 249, 250, and 251 are likely to be found in any sources. These isotopes, because of their low probability for spontaneous fission, contribute little to the overall neutron yield. However, their presence can influence the apparent half-life of the source when observed over long periods of time.

Gamma rays from the fission process and decay of fission products are also produced by californium neutron sources. The number of gamma ray photons per emitted neutron has been variously reported from about 2 to 5. The gamma ray spectrum is generally a continuum, with some 80% of the yield below 1 MeV.

Radioisotope (alpha,n) Sources

The use of alpha particles to create neutrons dates back to the discovery of the neutron by Chadwick in 1932. Here the first observed neutrons were produced by bombarding targets of boron or beryllium by alpha particles from the radioactive decay of polonium. Since that time, neutron sources based on the alpha particle irradiation of a suitable target have come to be the most

common type of laboratory scale neutron source based on radioactive decay.

TABLE 2 Characteristics of Be(alpha,n) Neutron Sources

Source	Half-life	E_α (MeV)	Neutron Yield per 10^6 Primary Alphas		Percent Yield with E_n<1.5 MeV	
			Calculated	Experimental	Calculated	Experimental
^{239}Pu/Be	24000y	5.14	65	57	11	9-33
^{210}Po/Be	138 days	5.30	73	69	13	12
^{238}Pu/Be	87.4 y	5.48	79[b]	--	--	--
^{241}Am/Be	433 y	5.48	82	70	14	15-23
^{244}Cm/Be	18 y	5.79	100[a]	--	18	29
^{242}Cm/Be	162 days	6.10	118	106	22	26
^{226}Ra/Be +daughters	1602 y	multiple	502	--	26	33-38
^{227}Ac/Be +daughters	21.6 y	multiple	702	--	28	38

[a]Does not include a 4 percent contribution from spontaneous fission of ^{244}Cm.

[b]From Ref. 2. All other data as calculated or cited in Ref. 3.

Table 2 shows some significant properties of alpha emitters often used in such sources. Ideally, the half-life of the alpha emitter should be neither too short nor too long. Practical demands on source life time require that the half-life be at least a few months. Half lives that are overly long, however, result in low specific activity of the material. High specific activity are preferred in order to minimize the mass of the material required for a given neutron yield.

A large number of these sources have been fabricated using ^{239}Pu as the alpha emitter. More recently, isotopes with a shorter half-life have come into favor. Foremost

among these is ^{241}Am, and americium-beryllium sources are now widely available. The apparent half-life of the neutron source may depend on the isotopic purity of the alpha emitter.

The most popular target material for (alpha,n) neutron sources is beryllium. Neutrons are produced through the reaction:

$$^{9}Be + \alpha \longrightarrow ^{12}C + n$$

The neutron yield for this reaction has been measured for thick beryllium targets, and is shown in Figure 1. When the alpha emitting material is finely dispersed throughout a beryllium matrix, yields very close to these values can be obtained. For typical alpha decay energies, only about 1 in 10^4 alpha particles results in a neutron-producing reaction before losing its kinetic energy. Therefore, relatively large activities of the alpha emitter are required to produce neutron yields of interest for practical neutron sources.

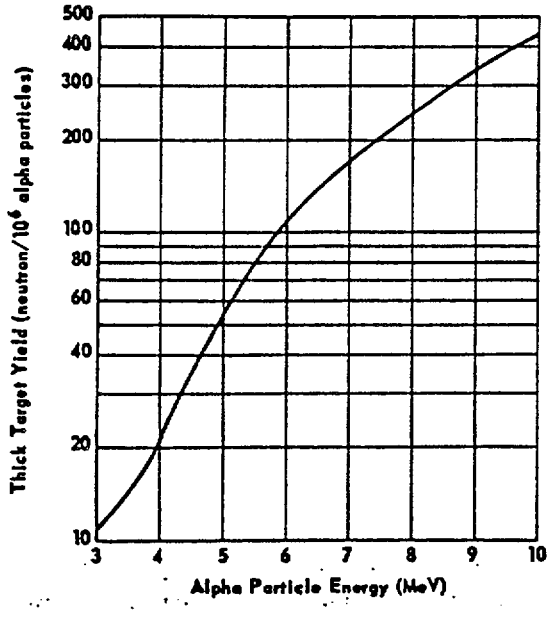

Fig. 1. Yield of neutrons from alpha particles incident on a thick beryllium target (from Ref. 2).

A common procedure in the fabrication of these sources is to form a metallic alloy between the alpha emitter and the host beryllium matrix. Actinides of the type shown in Table 1 will form an alloy with beryllium in the ratio of 1 actinide atom per 13 beryllium atoms. The alloyed mixture is then carefully encapsulated generally using double sealing techniques to guard against leakage of the large actinide activities.

The neutrons produced by typical alpha-Be sources cover a broad energy spectrum ranging from less than 1 MeV to greater than 12 MeV. This broad distribution is a result of two separate factors. The alpha particles, while nearly monoenergetic when formed, undergo continuous slowing down in the beryllium matrix, and may trigger a reaction at any point along their track. Secondly, neutrons from the reaction are observed at all angles with respect to the incoming alpha particle direction. Thus the kinematic spread in neutron energies contributes an additional broadening to the spectrum.

An example of a typical spectrum from an alpha-Be source is shown in Figure 2. This spectrum is for a

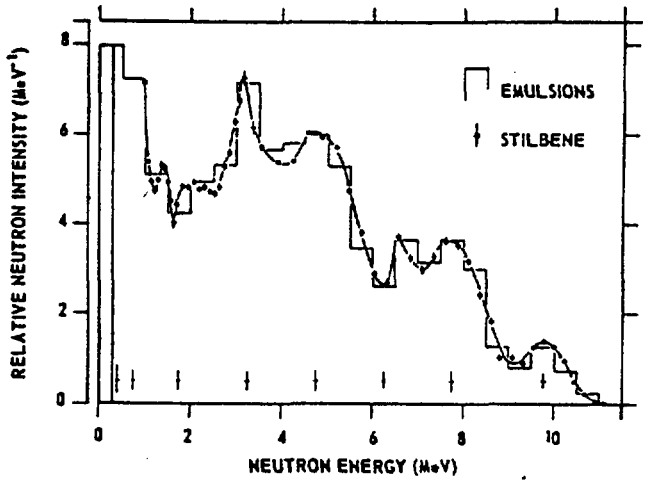

Fig. 2. A typical ^{239}Pu/Be neutron source spectrum (from Ref. 6).

source with small physical dimensions so that the neutrons emerge with a minimum of secondary interactions within the source. For large sources, however, it can be anticipated that additional reactions such as scattering, (n,2n) reactions, and possible fission events in the actinide component will alter the emitted neutron spectrum.

Although beryllium is by far the most common choice, other target materials are sometimes employed in conjunction with alpha emitters to form neutron sources. Some of these alternative target materials are shown in Table 3. In all cases the neutron yield is at least a factor of 5 below that of beryllium for equivalent alpha activity. The neutron spectrum produced varies considerably with the target material as illustrated in Figure 3. All of these spectra are considerably softer than that observed from a beryllium target. In some

TABLE 3 Alternative (α,n) Isotopic Neutron Sources

Target	Reaction	Q-Value	Neutron Yield per 10^6
Natural B	$^{10}B(\alpha,n)$	+1.07 MeV	13 for ^{241}Am alphas
	$^{11}B(\alpha,n)$	+0.158MeV	
F	$^{19}F(\alpha,n)$	-1.93 MeV	4.1 for ^{241}Am alphas
Isotopically Separated ^{13}C	$^{13}C(\alpha,n)$	+2.2 MeV	11 for ^{238}Pu alphas
Natural Li	$^{7}Li(\alpha,n)$	-2.79 MeV	
[For comparison:]			
Be	$^{9}Be(\alpha,n)$	+5.71 MeV	70 for ^{241}Am alphas

Data from Refs. 4 and 5.

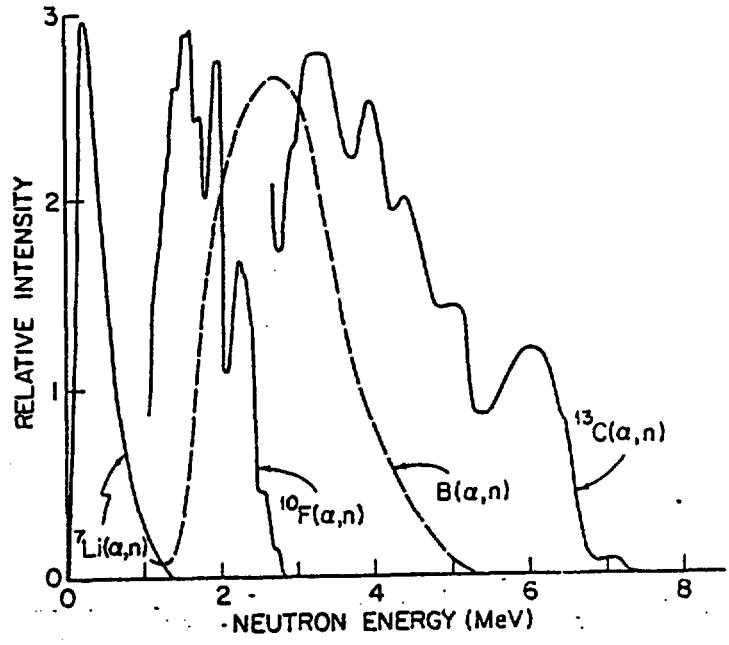

Fig. 3. Neutron spectra from several alternative (alpha,n) sources (data from Refs. 4 and 5).

applications, the lower average neutron energy can be an advantage that may offset the inherently lower neutron yield.

The neutron intensity obtainable from practical sources depends on the specific activity of the alpha emitter. With ^{239}Pu, yields of about 10^{6} neutron per second can be obtained from sources with physical dimensions of a few centimeters. Alpha emitters with greater specific activities such as ^{241}Am or ^{238}Pu can be used to fabricate small sources with yields ranging as high as 10^{8} to 10^{9} neutrons per second.

There are always gamma rays that accompany the neutrons emitted by such sources. Rather intense gamma ray backgrounds can be created through the decay of the alpha emitter in some cases, notably ^{226}Ra. In addition to these direct decay gamma rays, a small yield of gamma ray photons also may result from the neutron producing reaction. In beryllium, a 4.43 MeV gamma ray is produced

from the decay of an excited state in which the product ^{12}C nucleus can be left. The relative yield of this high energy photon is between .5 and .75 photons per neutron in typical alpha-Be sources (Refs. 7,8).

Photoneutron Sources

Neutrons can also be produced through the (gamma,n) reaction in light nuclei. The combination of a gamma ray emitting isotope and a suitable target material is therefore another alternative for portable neutron sources. As a practical matter, only deuterium and beryllium are used as targets through the following reactions:

	Q-value
$^{2}H + h\nu \longrightarrow {}^{1}H + n$	-2.226 MeV
$^{9}Be + h\nu \longrightarrow {}^{8}Be + n$	-1.666 MeV

All other possible targets nuclei require incoming gamma ray energies beyond those readily available from radioisotope sources.

The minimum energy required for these reactions to take place is just the absolute value of the reaction Q-value shown above. For gamma rays with higher energy, neutrons are produced with an angle dependent energy given by the following relation:

$$E_n(\theta) = \frac{M(E+Q) - E_\gamma^2/2}{m+M} + \frac{mE_\gamma^2\cos^2\theta}{(m+M)^2}$$

$$+ \frac{E_\gamma\cos\theta}{(m+M)^2}\left\{(2mM)(m+M)(E_\gamma+Q) - (E_\gamma^2)(mM+m^2\sin^2\theta)\right\}^{1/2}$$

where θ = angle between gamma photon and neutron direction
E_γ = gamma ray energy
M = mass of recoil nucleus x c^2
m = mass of neutron x c^2

While the above relation predicts a range of neutron energies for a given gamma ray photon energy, the variation with angle is only a few percent of the mean neutron energy. Therefore, a unique feature of photoneutron sources is the nearly monoenergetic nature of the emitted neutrons, provided only one gamma ray above threshold is present. Furthermore, using commonly available radioisotope gamma ray sources, neutrons are produced with typical energies in the 10-1000 keV range as opposed to the much higher average energies typical of spontaneous fission or alpha-Be sources.

Figure 4 shows one possible source configuration. In this case a spherical gamma ray emitting core is surrounded by a shell of either beryllium or deuterated polyethylene. The spherical symmetry of the source

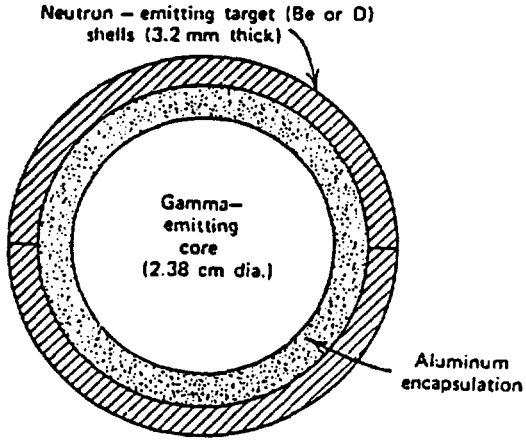

Fig. 4. A specific example of a spherical photoneutron source. The source dimensions correspond to the yield figures given in Table 4.

TABLE 4 Photoneutron Source Characteristics

Gamma Ray Emitter	Half-Life*	Gamma Energy (MeV)*	Target	Neutron Energy (keV)**	Neutron Yield, n/s for 10^{-10} Bq activity***
^{24}Na	15.0 h	2.7541	Be	967	340 000
		2.7541	D	263	330 000
^{28}Al	2.24 m	1.7787	Be	101	32 600
^{38}Cl	37.3 m	2.1676	Be	446	43 100
^{56}Mn	2.58 h	1.8107	Be	129	91 500
		2.1131		398	
		2.9598		1149	
		2.9598	D	365	162
^{72}Ga	14.1 h	1.8611	Be	174	64 900
		2.2016		476	
		2.5077		748	
		2.5077	D	140	25 100
^{76}As	26.3 h	1.7877	Be	109	3 050
		2.0963		383	
^{88}Y	107 d	1.8361	Be	152	229 000
		2.7340		949	
		2.7340	D	253	160
^{116m}In	54.1 m	2.1121	Be	397	15 600
^{124}Sb	60.2 d	1.6910	Be	23	210 000
^{140}La	40.3 h	2.5217	Be	760	10 200
		2.5217	D	147	6 600
^{144}Pr	17.3 m	2.1856	Be	462	690

* Decay data from C. M. Lederer and V. S. Shirley, Table of Isotopes, 7th Edition, Wiley Interscience, NY (1978)

** Calculated for $\theta = \pi/2$, approximate midpoint of primary spectrum

***Monte Carlo calculations for the source dimensions given in Fig. 6. outer target shells are either metallic Be or deuterated polyethylene. Core materials assumed to be NaF, Al, CCl_4, M_nO_2, Ga_2O_3, As_2O_3, Y_2O_3, In, Sb, La_2O_3, and Pr_2O_3.

assures an isotropic yield of neutrons. Other configurations with widely varying geometries have been used in various applications. In some cases the gamma ray emitting source is separable from the target material to allow "switching off" the source neutrons when desired.

Table 4 lists the common gamma ray emitters used for photoneutron sources and the resulting properties of the neutrons produced from either beryllium or deuterium. The yields that are shown in the last column are for the physical dimensions shown in Figure 4. Greater yields can be achieved by increasing the target shell thickness, but only at the expense of increased scattering that tends to degrade the monoenergetic nature of the neutron spectrum. Most of these gamma ray sources have short half-lives so that their use is only practical near a production facility such as a research reactor. One notable exception is the antimony-beryllium source with a 60 day half-life. This type of photoneutron source has been used in a significant number of remote applications.

Some typical energy spectra for photoneutron sources are shown in Figure 5. Each spectrum consists of two components: a primary peak whose width is determined by the reaction kinematics, and a low energy "tail" that results from neutron scattering within the source itself. It is this latter component that becomes a more significant component as the source dimensions increase. The spectra shown were obtained by Monte Carlo calculations for the dimensions shown in Figure 4.

The major disadvantage of photoneutron sources is their very intense gamma ray background. For typical

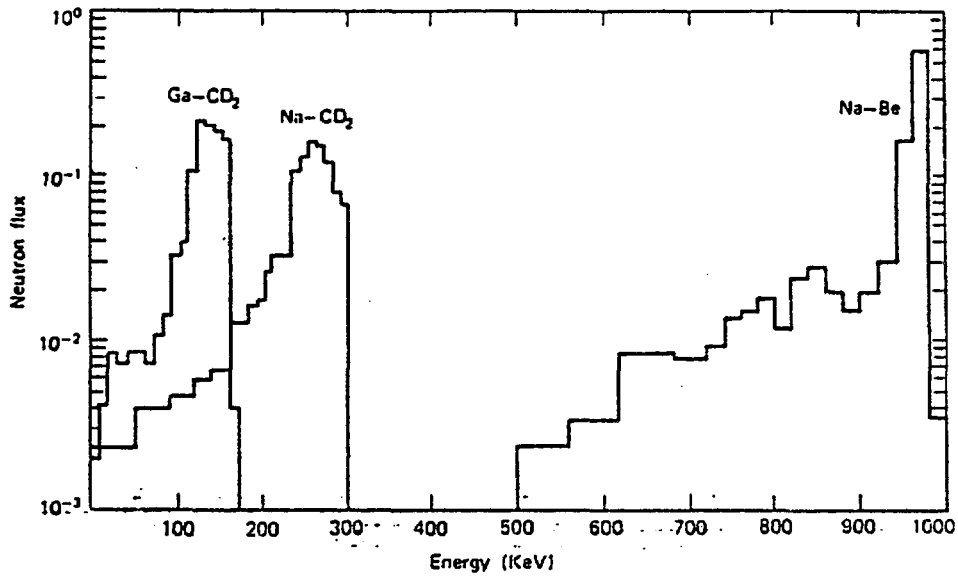

Fig. 5. Calculated neutron spectra for three different photoneutron sources with dimensions given in Fig. 4. The gamma emitters are either ^{72}Ga or ^{24}Na. The outer shells are either deuterated polyethylene (CD_2) or beryllium.

sources, the gamma-to-neutron ratio is on the order of 10^5. Thus many measurements involving gamma ray sensitive detectors are made difficult or impossible. In some applications, these problems can be alleviated by surrounding the source with a thick shield of lead or other heavy material. The gamma ray flux can thereby be greatly reduced, but the additional neutron scattering created by the presence of the shield degrades the neutron spectrum into a broad continuum with average energies significantly below those shown in Figure 5.

Intercomparisons and Applications

Probably the most common application of radioisotope neutron sources is in the calibration of instruments, particularly those used in radiation protection measurements. In these cases, photoneutron sources are generally not acceptable because of their high gamma ray background. Either californium sources or alpha-Be

sources are therefore more common in these applications. It is often of critical interest to know the details of the energy distributions of the neutrons in these cases, and much of the recent effort at calculating and measuring these spectra has been motivated by this need.

Radioisotope sources have also been used widely in neutron physics experiments. The fission spectrum available from californium sources approximates the fission neutron spectrum expected from common fissionable isotopes. Reaction rate measurements using a californium source therefore closely match those expected in other unmoderated fission spectra. The monoenergetic yield from photoneutron sources is of great advantage in energy differential cross section measurements. At the University of Michigan, we have exploited such sources for a wide variety of fission and other neutron induced reaction cross section measurements.

Radioisotope sources are also in common use as startup sources in nuclear reactors. An antimony-beryllium source serves nicely in this capacity, since the antimony activity is easily replenished during full power operation of the reactor.

Isotope sources have also been widely used in geophysical measurements. While their use in borehole logging is now being gradually replaced by compact neutron generators of the D-T type, they have proved their usefulness in analysis of materials through neutron moderation and capture gamma ray analysis.

Radioisotope neutron sources also find application in standards measurements. The primary standard neutron

source in the United States (NBS-1) is a spherical radium-beryllium photoneutron source maintained by the National Bureau of Standards. This configuration is felt to have a high degree of long term stability necessary for standards applications.

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TWO EXAMPLES OF CYCLOTRONS IN BASIC RESEARCH AND MEDICAL APPLICATIONS

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Abstract

After a brief examination of the chronological evolution of cyclotrons, two current designs of such machines used as research tools will be reviewed.

The first, deals with the design of a cyclotron for basic research in nuclear physics for the 1990's. Required performances, choices of the cyclotron parameters and proposed technical solutions will be presented on the basis of the Orsay Superconducting Cyclotron project.

The second, concerns the applied research, more precisely the medical applications with the example of MEDICYC which is a medical cyclotron programme undertaken by the Nice Cancer Centre. Its major goal is radiotherapy based on neutrons and protons but this machine should also satisfy the medical requirements for the Nuclear Medicine Department of the Hospital.

Economic considerations of both cyclotrons will be shortly discussed.

I. CYCLOTRONS AND THEIR EVOLUTION

The 1984 Tenth International Conference on Cyclotrons and their applications was held from April 30th to May 3rd in East-Lansing (Michigan), organised by the National Superconducting Cyclotron Laboratory which has built the pionnering cryogenic cyclotron. The purpose of these conferences is to meet accelerator physicists, engineers and also the increasing number of those concerned with applications of cyclotrons.

The field of applications of cyclotrons is now wide due to the fact that nuclear physics is a discipline which has produced a broad impact on our present-day society: radioisotopes are extensively used in medicine, geology, analytical chemistry and numerous industrial applications.

To understand the development of cyclotrons it is necessary to follow the chronological evolution of these accelerators in the nuclear physics research field.

In the early 1930's two important events occured : one in Europe, in the Cavendish Laboratory, where Lord Rutherford and two young scientists, Cockroft and Walton produced the first artificial nuclear reaction Li^7 (p,α)α using a proton beam of 125 kV from an electrostatic machine. This kind of accelerator has known a great development and "tandems" have now attained noteworthy performances.

The other event occured in the United States at about the same time in California where E. Lawrence developped the cyclotron, a machine which has known also a remarkable development. Accelerating ions of mass m and charge state q in a static magnetic field B requires the meeting of four main functions which make a cyclotron:

- Injection of these ions in the central region of the median plane of a magnet (from any internal or external device which, of course, includes the ion source).

- Guiding and Focusing these ions by an adequate magnetic field, in a solid pole magnet or split pole magnet.

- Acceleration of the ions with an HF-voltage on cavity working with a frequency related to the ions frequency :

$$\omega_{HF} = h\,\omega_{ion} = h\,\frac{qB}{m} \qquad \text{where } h = 1,2,\ldots \text{ etc.}$$

- Extraction of the ions on the edge of the magnet where they have reached their maximum energy by a system which delivers the beam in the user area.

The solutions for solving these functions have given different cyclotron families. These developments were necessary to satisfy mainly three requirements of nuclear physicists :

- The main requirement is certainly a precise knowledge, and that means control, of beam characteristics on the target. This concerns the emittance, the time structure, the absolute energy and the spread in energy.

- Another important requirement is the possibility to change the beam energy and the type of accelerated projectile, inevitably of course as fast as possible.

- A final requirement leads to new big cyclotrons. It concerns the final energy which is always going higher. (The GeV energy range for protons is planned ...).

Hence, the "working horse" for the cyclotron development has always been the nuclear physicist. His two first requirements are for cyclotrons becoming so precise and so versatile that they will be comparable to electrostatic machines (tandems) . The third requirement of the physicist which has led to big split magnet machine, can be fulfill only by cyclotrons, linacs or fast cycling synchrotrons.

Many cyclotron facilities have important applications programmes and significant fractions of the operating time, even on the largest machine (SIN for example) are devoted to medical applications. In fact these applications started quite early (the brother of Ernest, John Lawrence was biologist!) but always with machines which have not been designed for medical purposes. In fact we are just entering the dedicated medical cyclotron era. For this field, the first requirement of the physicist although slightly less stringent is for an increased reliability, in particular for radiotherapy, where a cheaper overall cost of the machine is requested.

The table I presents a short, but not exhaustive, survey of about 50 years of cyclotron development.

Table I - Chronological Evolution of Cyclotron and milestones events

1930	E. Lawrence (Nobel Prize 1939). His brother John was a biologist and used the cyclotrons for neutron production ...
1938	Thomas : Azimuthal varying Field Cyclotrons (Strong focusing principle).
1939	Cyclotron No.7 in Berkeley (1.50 m. pole diameter)
1950	Classical Lawrence cyclotrons and modulated cyclotrons (synchro-cyclotrons).
	Pioneering sector-focused cyclotron at Delft.
1958	CERN: Synchro-cyclotron (600 MeV-protons, 5 m pole diameter)
1959	Sea Iasland Conference on "Sector Focused Cyclotrons"
1970	First idea on ring cyclotrons (i.e. "Separation of functions guiding-focusing (magnet) from acceleration (RF cavities).
	Second generation of big cyclotrons (SIN-TRIUMF)
	First industrial realisations of compact cyclotrons
1974	(18 January) a 590 MeV proton beam emerged from the SIN-Ring
1975	Design studies for a National Heavy ions Laboratory (GANIL) based on 3 coupled cyclotrons.
	First studies on superconducting cyclotrons in Michigan and Milan.
1982	First beam out at Michigan.

II. AN EXAMPLE OF CYCLOTRON DESIGN FOR 1990's NUCLEAR PHYSICS[1]

In the 60's to 70's many laboratories built cyclotrons for basic nuclear physics research. These machines accelerated light ions (i.e. protons, deuterons, α) to energies below 100 MeV. In the intervening years since new trends appeared in this science leading to different fields.

a) The heavy ions era :

Acceleration of ions covering the full list of the Mendeleev's classification. This era of "heavy ions" may be decomposed in two periods :

- A first period (1970-1980) which could be called "conversion or boosting of existing cyclotrons", deals with acceleration of masses between 12 and 40 in the energy range from a few MeV per nucleon to about 15 MeV per nucleon. This physics field covered mainly three points:
  - study of fast rotating nuclei
  - dynamics of nucleus-nucleus collisions, looking for new mechanisms
  - production and study of nuclei far from stability.

An extension of these research lines towards higher energies appears at the end of the 70's.

- A second step which could be called "period of coupled machines" is now opening for the 1980-1990 years. The table II gives examples where cyclotrons are involved (of course, this concerns other types of accelerators (tandem + linac, linac + synchrotrons)).

TABLE II - Examples of coupled machines

CYCLOTRON+	*CYCLOTRON OR TANDEM*	
EXAMPLES OF FACILITIES	*MSU (USA)*	*VICKSI (FRG)*
	GANIL (F)	*OAK-RIDGE (USA)*
	SARA (F)	*CHALK-RIVER (CANADA)*

b) The intermediate energy physics

This field leads to higher energy, light particle accelerators where cyclotrons (SIN (Switzerland), TRIUMF (Canada)) play an active role together with other accelerators (LINAC and synchrotron).

Satisfying the requirements for both fields has led to a new kind of cyclotron, where the magnet is split in sectors. This was the first important step towards the separation of the functions of cyclotrons: this successfull principle has given important improvements of performances (in quantity as well as in quality!)

- higher intensity and energy due to the increased vertical magnetic focusing

- higher energy spread of the beam due to the single turn extraction which was made possible by the introduction of large RF accelerating cavities between the magnet sectors.

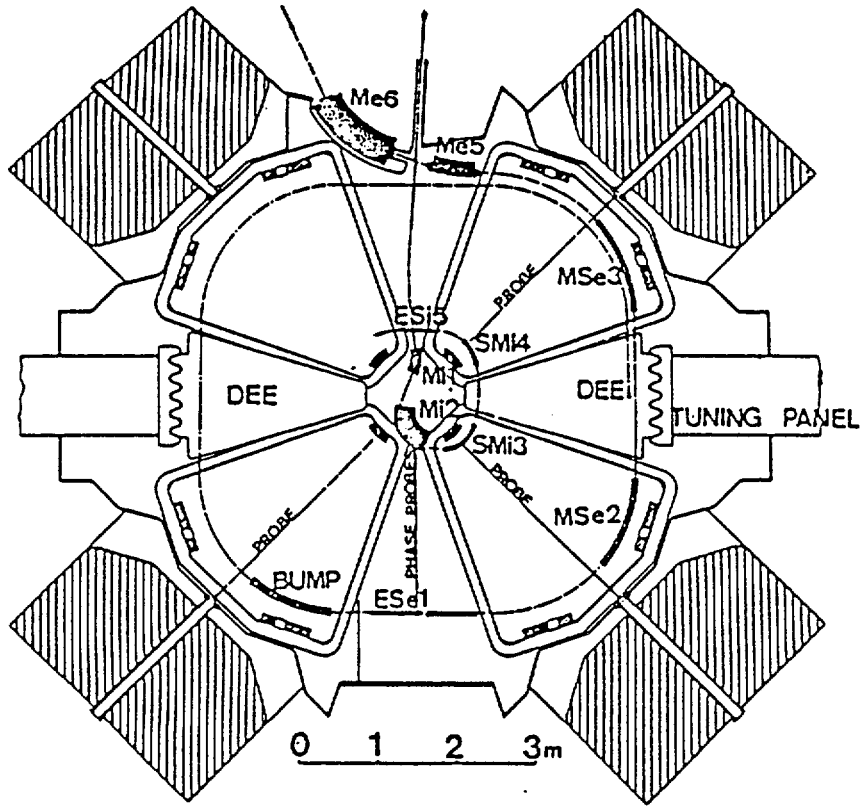


FIG. 1. - Horizontal cross section of a separed sector cyclotron (GANIL)

However, these machines have a reduced average field, this means a bigger average final radius for a given maximum energy and this leads of course to rather ambitious and expensive research programmes. The question arises: Is there anything left in the nuclear physics field?

c) The example of the Orsay Superconducting Cyclotron programme

Many nuclear physicists are interested in the field of nuclear structure and nucleons inside the nucleus. Such studies can be made with the required accuracy by using light ions probes. It is evident that the number of laboratories involved in such research is far less than is appropriate for the magnitude of the tasks. Recent progress demonstrates the need for an ideal facility which would give the performances summarized in Table III.

TABLE III - Requirements for an advanced light ions facility

ENERGY RANGE : . 50 TO ABOUT 300-400 MEV PROTONS
. UP TO 150 MEV/NUCLEON FOR D, α, LI, C
HIGH ENERGY RESOLUTION : ABOUT 5 X 10^{-4} OR BETTER!
SMALL EMITTANCE : ABOUT π MMXMRAD (CF. TANDEM!)
GOOD INTENSITY : 10^{12} - 10^{14} PPS
FLEXIBLE DUTY-CYCLE : MACRO D.C. 100%
MICRO (< 0.5 NS) 1% TO 20%
POLARIZED ION SOURCE : P, D, LI

Moreover, on the heavy-ions front, with the advent of highly stripped heavy ions sources such as the electron cyclotron resonance (ECR)[2] ion source or electron beam ion source (EBIS)[3] a pre-stripper accelerator stage for coupled heavy ions machines has become less necessary and injection of these ions in a single machine is regarded today as an attractive and less expensive way for nuclear physics research.

Nevertheless the need for high energies remain which means that high B^2R^2 is necessary remembering that R is always expensive (big cyclotron diameter means expensive machine and expensive buildings to house it).

These two arguments, new sources and high B, have led to the choice of compact cryogenic cyclotron and axial injection of highly stripped ions at low energy[4].

The Table IV presents the main characteristics of the Orsay project of a superconducting cyclotron for an ion of charge state Zi and mass A. The focusing constant KF gives the energy which can be obtained for light ions ($E = K_F \frac{Z_i}{A}$) and the focusing constant KB the energy for heavy ions ($E = K_B \frac{Z_i^2}{A^2}$)

Table IV - Main characteristics of the Orsay Project of a superconducting cyclotron.

$K_B = 600$, $T_{max} = K_B \frac{Z_i^2}{A}$ for $\frac{Z_i}{A} \leq 0.37$

$K_B = 111$

$K_F = 222$, $T_{max} = K_F \frac{Z_i}{A}$ for $\frac{Z_i}{A} > 0.37$

3 magnetic sectors

3 electrical dees opening 60°

Average magnetic field : $\overline{B} = 1.75$ to 4.05 T

Extraction radius : 0.87 m

R.F range : 24 to 62 MHz

Harmonic modes : 2, 3, 4

Dee voltage peak : 100 kV

No internal source

Axial and radial injection

The characteristics of some typical ions injected from an external source are reported in table V (ions heavier than carbon could also be radially injected from the existing Orsay MP Tandem, which gives more flexibility to this installation).

Table V - Characteristics of some typical ions accelerated in the Orsay project from an external source.

h	ion	$\frac{Z_i}{A}$	Energy (MeV/A)	B_o (Tesla)	V (kV)
2[a]	proton	0.9928	207.4	2.04	62.2
2[a]	$^{3}He^{++}$	0.6634	130.1	2.55	98.0
2[b]	$^{20}Ne^{7+}$	0.35	63.	3.55	100.0
3[b]	$^{40}A^{9+}$	0.23	27.	3.65	76.0
4[b]	$^{84}Kr^{10+}$	0.12	8.	3.87	66.3
4[b]	$^{131}Xe^{17+}$	0.13	9.5	3.89	73.0

a) from duoplasmatron
b) from ECR

Figure 2 shows the energy range versus Zi/A diagram taking into account focusing limitations, frequency range and inflector electric field limitations for the axial injection.

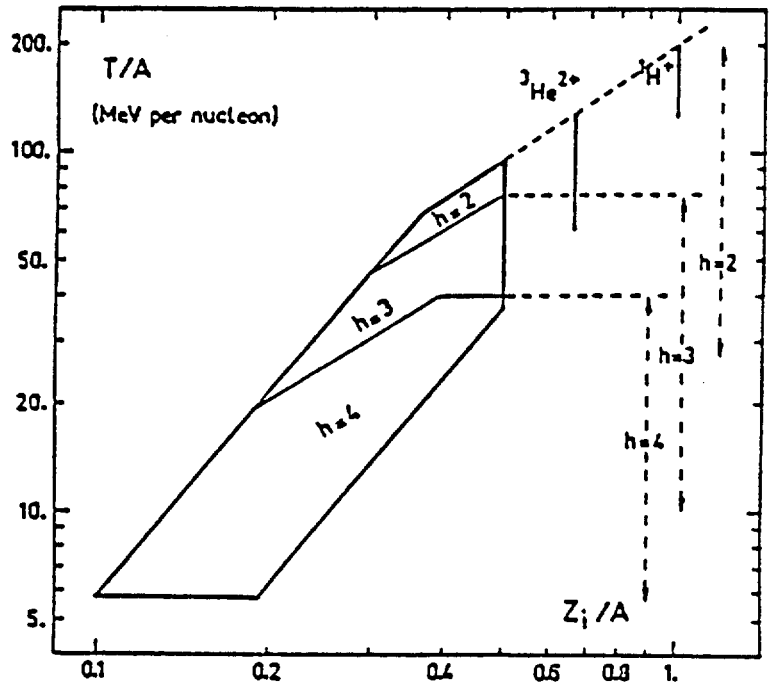


FIG. 2. T/A, Zi/A diagram for the Orsay project

Figures 3, 4 and 5 show a schematic view and two cross sections of this machine with the relevant dimensions.

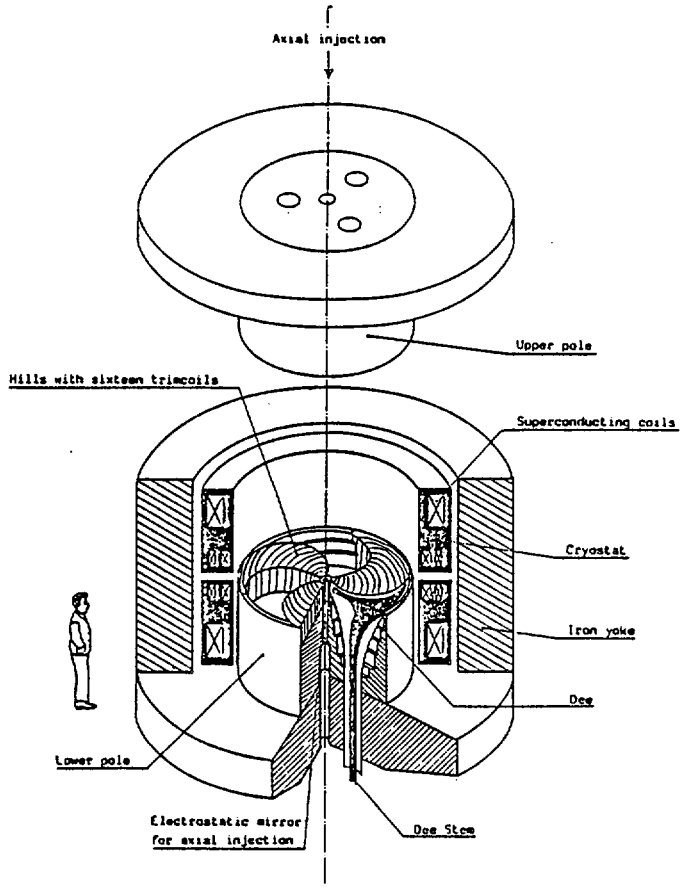


FIG. 3. Exploded view of the Orsay cryogenic Cyclotron

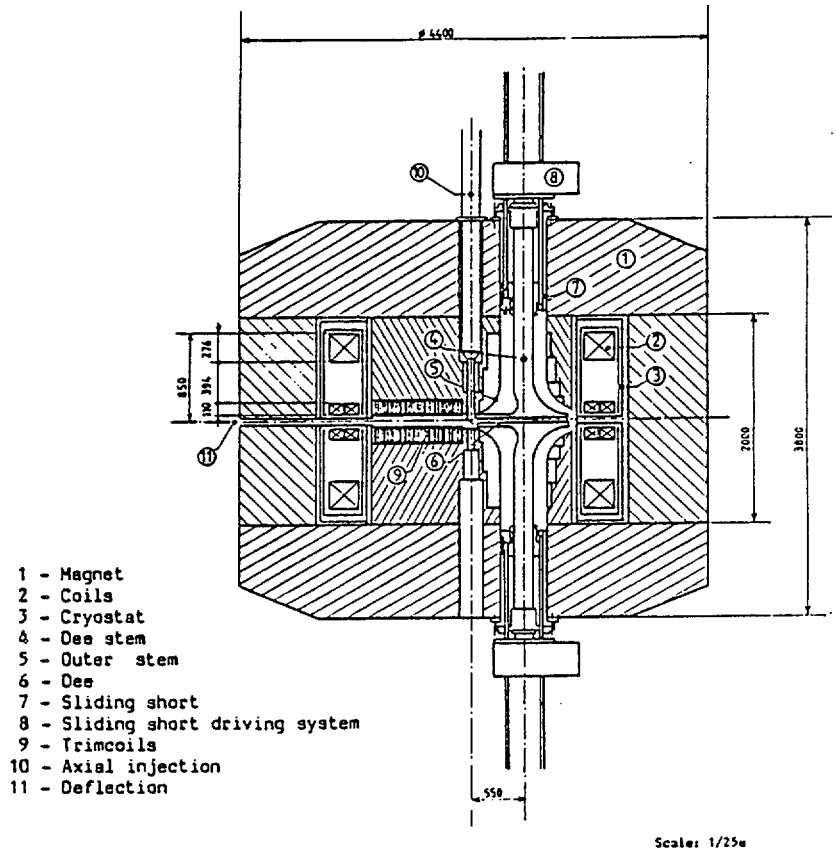


FIG. 4. Vertical Section of the K600 Cyclotron

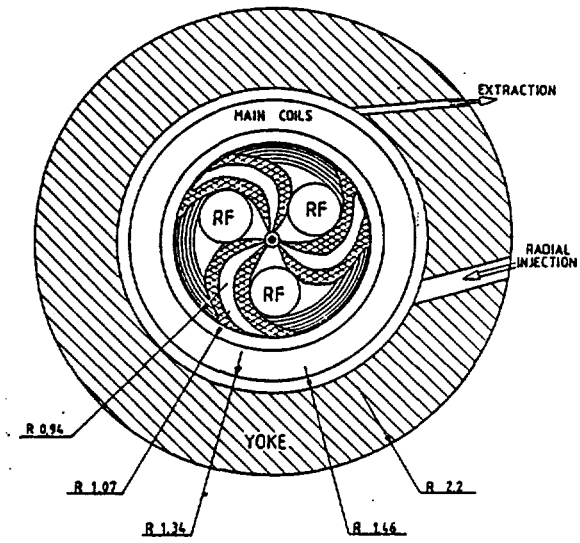


FIG. 5. HORIZONTAL CROSS SECTION

III. THE EXAMPLE OF THE MEDICYC[5] MEDICAL CYCLOTRON

The Nice cancer Center has undertaken a medical cyclotron programme whose major goal is radiotherapy based on fast neutrons and, in particular cases, on protons. Taking into account the high cost of such a machine, the possibility of producing radio-isotopes most commonly used in a hospital has been also considered. The figure 6 presents a schematic view of the planned installation.

a) The radiotherapy programme

Fast neutrons were first applied for therapeutic purposes by Stone[6] in Berkeley in 1938, but due to the lack of data in biology (the R.B.E. of neutrons was not known!), wrong doses have been applied and late complications appeared which justified the end of this programme. In 1969, neutrontherapy was initiated at the Hammersmith hospital in London[7] after extensive radiobiological investigations. First encouraging results appeared but were not confirmed in other centres, due to the difficulties of comparison between clinical trials (different cyclotrons, different target, different irradiation fields, etc.). However, taking into account recently published data, there appears to be evidence of a benefit of neutrons for

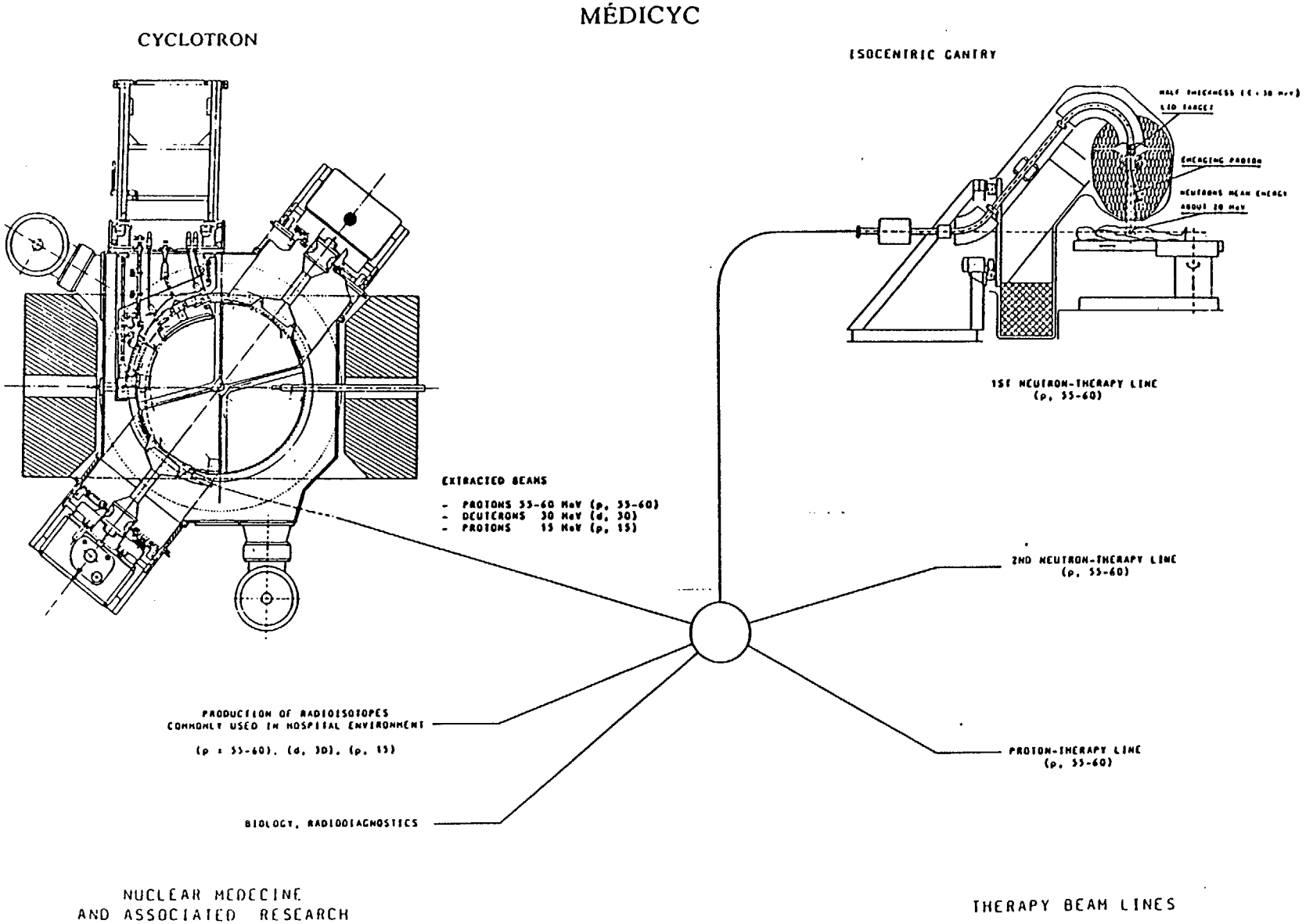

MÉDICYC
CYCLOTRON
ISOCENTRIC GANTRY
EXTRACTED BEAMS
- PROTONS 55-60 MeV (p, 55-60)
- DEUTERONS 30 MeV (d, 30)
- PROTONS 15 MeV (p, 15)
1ST NEUTRON-THERAPY LINE
(p, 55-60)
2ND NEUTRON-THERAPY LINE
(p, 55-60)
PROTON-THERAPY LINE
(p, 55-60)
PRODUCTION OF RADIOISOTOPES
COMMONLY USED IN HOSPITAL ENVIRONMENT
(p : 55-60), (d, 30), (p, 15)
BIOLOGY, RADIODIAGNOSTICS
NUCLEAR MEDECINE
AND ASSOCIATED RESEARCH
THERAPY BEAM LINES

FIG. 6.

some tumours types and characteristics of a dedicated cyclotron can be defined as shown in the column Neutrontherapy of Table VI.

TABLE VI - Medical requirements and operating parameters.

MEDICAL REQUIREMENTS	NEUTRONTHERAPY	PROTONTHERAPY	NUCLEAR MEDECINE
	NEUTRONS . MEAN ENERGY 20 MEV . DOSE RATE 3.36cGy/MIN . 50% DOSE D>14 CM	PROTONS . ENERGY 60 MEV	$^{11}C.\ ^{13}N.\ ^{15}O.\ ^{18}F.\ ^{123}I$
CYCLOTRON BEAMS	PROTONS 60 MEV INTENSITY I = 15 uA	PROTONS 60 MEV	PROTONS 60 MEV DEUTONS 30 MEV PROTONS 15 MEV FROM H2
OPERATING HARMONIC MODES	FONDAMENTAL	FONDAMENTAL	FONDAMENTAL AND SECOND HARMONIC

A cyclotron accelerating protons at 60 MeV, associated with a new target (Lid) has been chosen for this purpose.

Following the Harvard Cyclotron Laboratory[8] who have successfully treated ocular malignant melanoma (the tumor control rate is close to 100%) with a 60 MeV-0.8nA proton beam, this particular application of the direct proton beam has been included in the medical programme.

b) The radio-isotopes programme

This machine has been designed in such a way that by changing the magnetic field, still with the same RF frequency (24MHz) it is possible to accelerate other ions, on different harmonic modes, for the production of isotopes for the nuclear medicine department.

I^{123} is produced at high energy, by the simple (p,5n) reaction on the fundamental mode (h=1) with the 60 MeV proton beam.

The second harmonic mode (h=2) allows the acceleration of deuterons at 30 MeV, which is a very convenient energy for F^{18} production from ^{20}Ne in NTP conditions.

The same harmonic mode can be used for acceleration of H_2^+ which is very abundant in duoplasmatron sources. From these ions we get 15 MeV protons for production of ^{15}O, ^{11}C and ^{13}N.

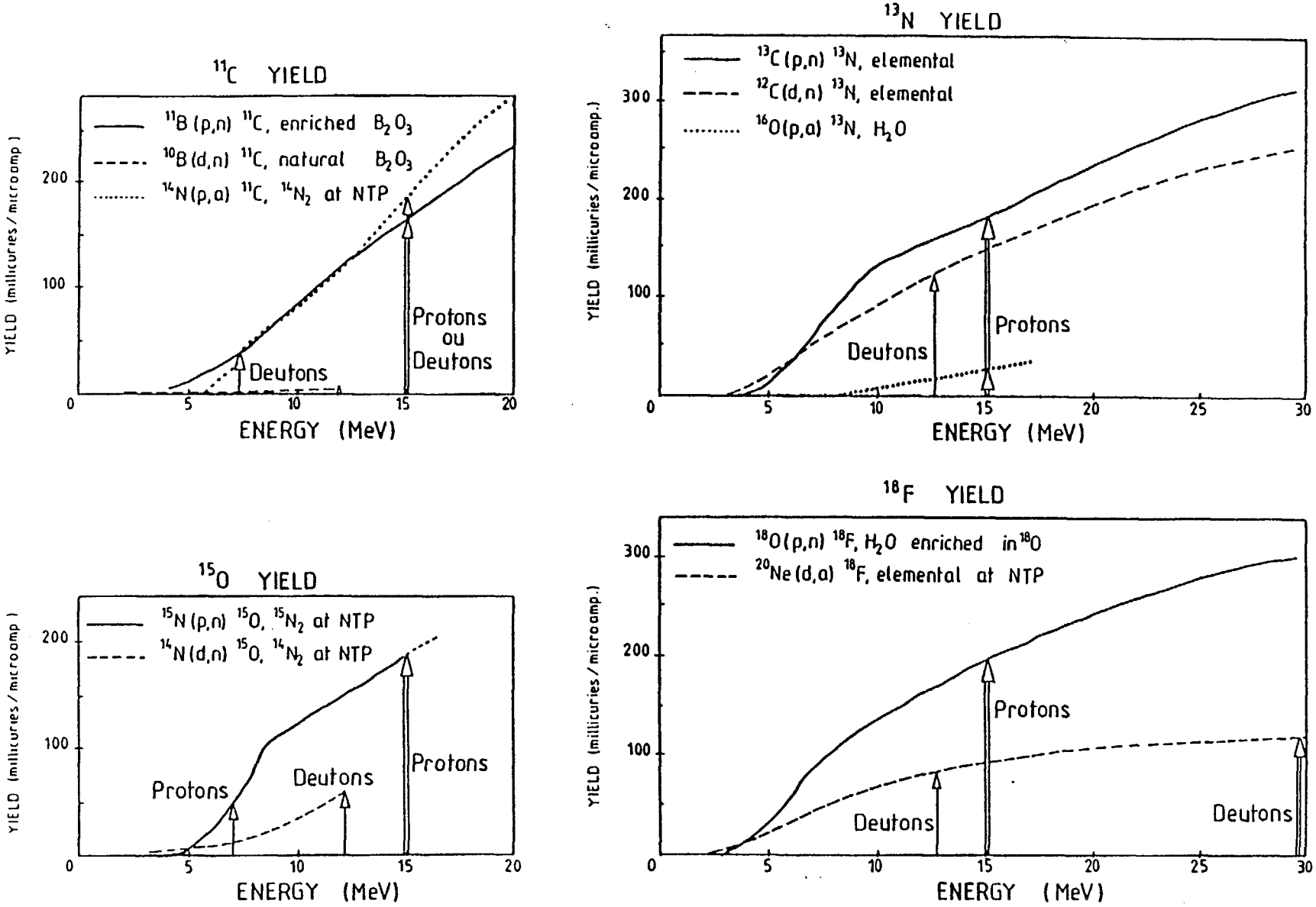


FIG. 7. Radioisotopes production on second harmonic (═══) and third harmonic (——).

The yields of these reactions are presented on the figure 7. A third harmonic (h=3) acceleration mode is also possible with this cyclotron because a 75° Dee aperture angle has been chosen. This gives a less expensive way for producing ^{15}O, and ^{13}N by (d,n) reactions for two reasons : the cyclotron is operated at a lower magnetic field and the targets are simpler.

c) The technical choices of the cyclotron

A fixed frequency cyclotron producing discrete energies for different ions axially injected from an external duoplasmatron source has been chosen. this machine is now in construction. The characteristics are summarized in the following table :

TABLE VII - Characteristics

CHARACTERISTICS
. MAGNET LENGTH : 4.0M HEIGTH : 2.3M WIDTH : 1.7M SECTORS : 4 TRIMCOILS : 10
. INJECTION EXTERNAL SOURCE : DUOPLASMATRON BUNCHING : SINGLE HARMONIC BUNCHER FOCUSING : 2 MAGNETIC CYLINDRICAL LENSES INFLECTOR: SPIRAL TYPE.
. RADIOFREQUENCY SYSTEM 2 DEES OF 75° APERTURE FREQUENCY : 24 MHZ PEAK VOLTAGE : 50 KVOLTS FOR H=1 30 KVOLTS FOR H=2 POWER : 2 CW 25000 EIMAC
. EJECTION FIRST SECTION : ELECTROSTATIC E=100 KVOLTS/CM SECOND SECTION : ELECTROMAGNETIC B=2500 GAUSS G=+400 G/CM THIRD SECTION : 1 MAGNETOSTATIC CHANNEL = + 1000 G/CM

1. The radiofrequency system

On one hand a fixed frequency RF system is more simple and cheaper than a variable frequency one. There are no sliding contacts in vacuum or air, no big cavities, no broad-band amplifier and neutralization. The driving amplifier can be at relatively low power

and the whole control chain is greatly simplified. On the other hand, chosing 75 degrees angle for both dees, which can be fed at 0 or 180 degrees, allows acceleration in three harmonic modes.

2. Axial injection

A simple axial injection system has been studied. It presents many advantages. Just the simple fact to put the source outside the machine simplifies all the problems involved:

α) We do not have space problems despite using a cumbersome duoplasmatron source giving a better emittance (normalized emittance 1.5 π mmxmrad) than the usual internal one.

β) We can utilize an external buncher (about 400 volts).

γ) We have a tremendous vacuum improvement : only 1000 l/sec are necessary to obtain 10^{-6} Torr in the machine.

δ) The source can be located immediately above the magnet (figure 8). The injection optics are simplified to two cylindrical magnetic lenses and two diaphragms (Figure 9). The unwanted ions are reduced at the inflector entrance to about 2,5%.

ε) The size of the inflector can be reduced allowing it to be located inside the dummy-dee minimising the dee-to-dee capacitance.

ζ) Because of this arrangement a wider RF gap is made available which allows higher accelerating voltages (> 50 Kilovolts) and safer operating.

η) A single central system geometry [4] has been found allowing on axis injection and constant geometrical set-up compatible with the various harmonic modes (figure 10).

θ) Finally, because of α) and β) a separated turns extraction seems quite feasible and that is very important for the reliability of a medical machine.

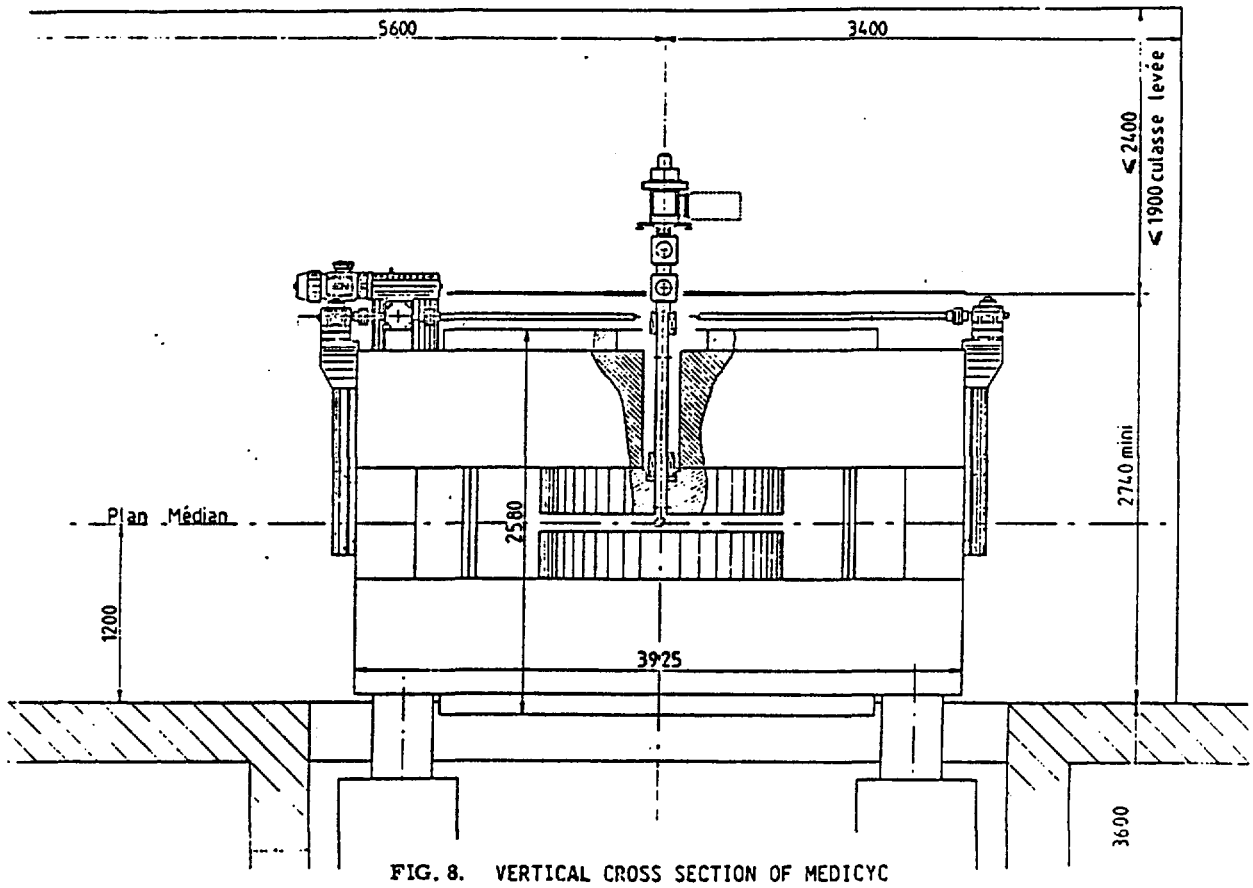


FIG. 8. VERTICAL CROSS SECTION OF MEDICYC

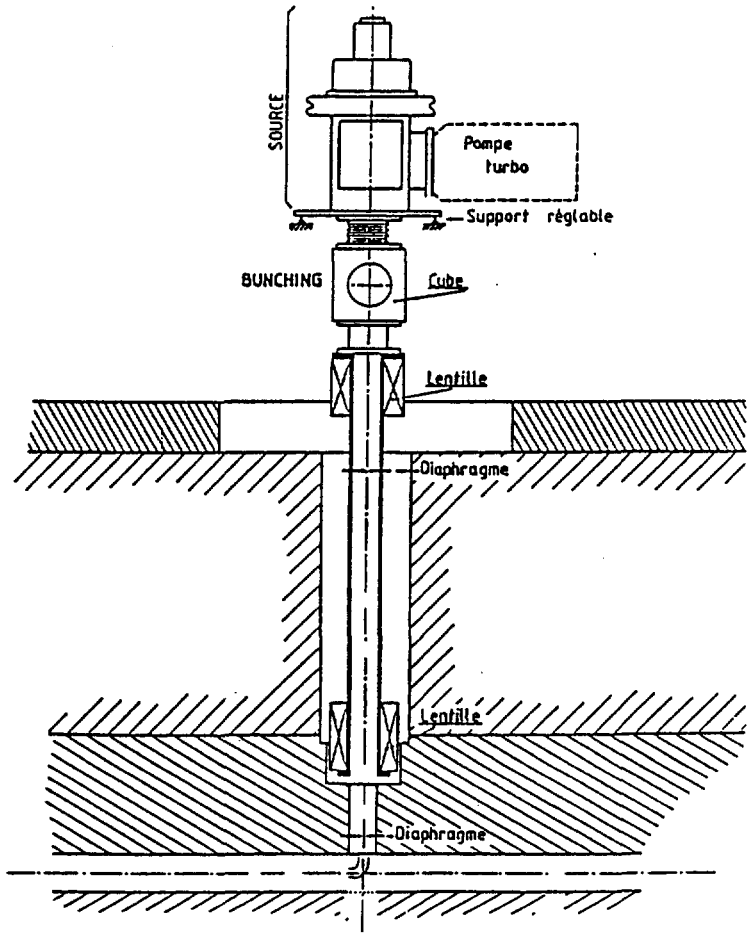


FIG. 9. MEDICYC axial injection line

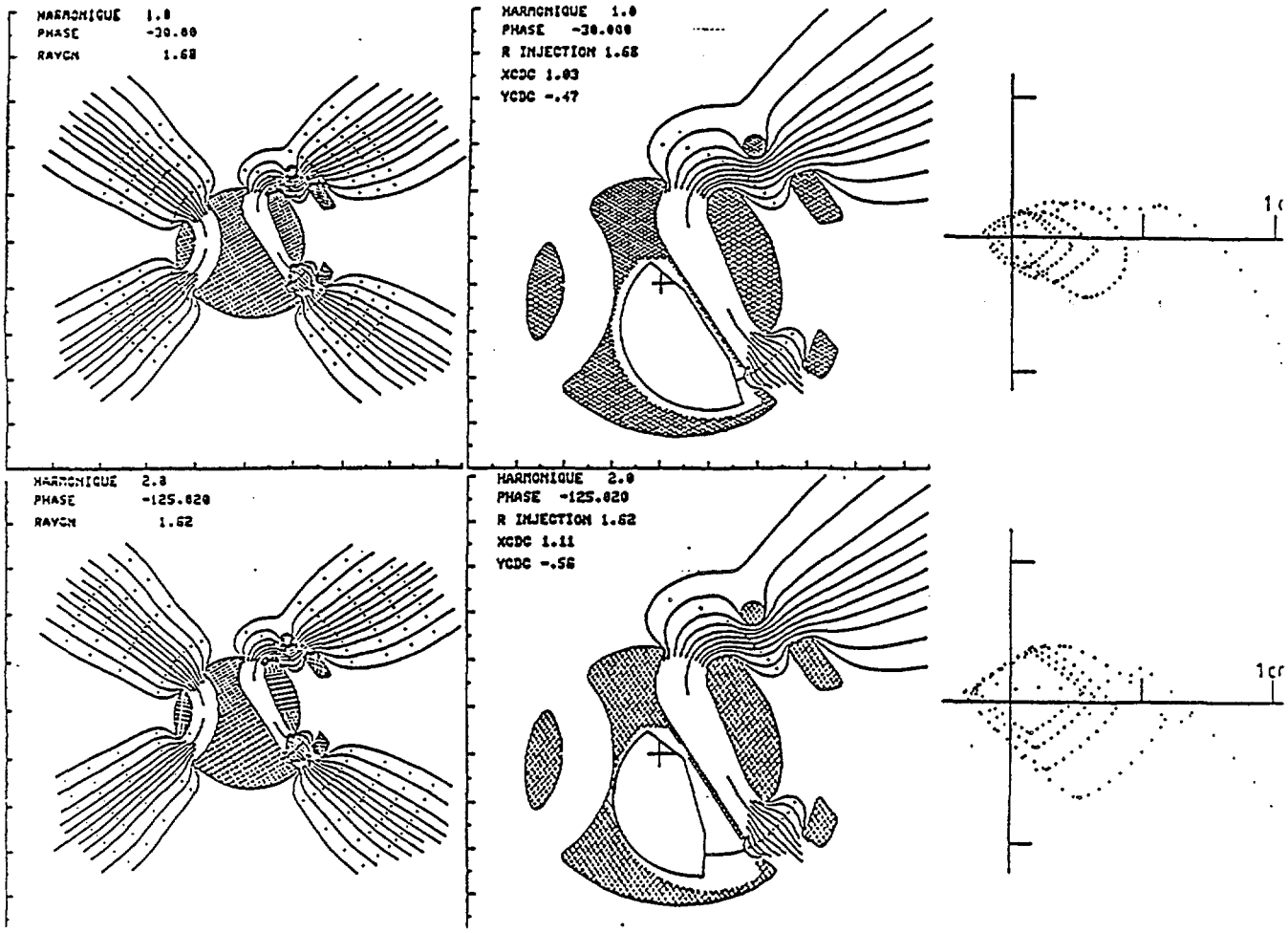


FIG. 10.

IV. ECONOMIC CONSIDERATIONS OF BOTH CYCLOTRONS

a) Estimated costs for the Orsay project

The development of the costs are summarized in the following table :

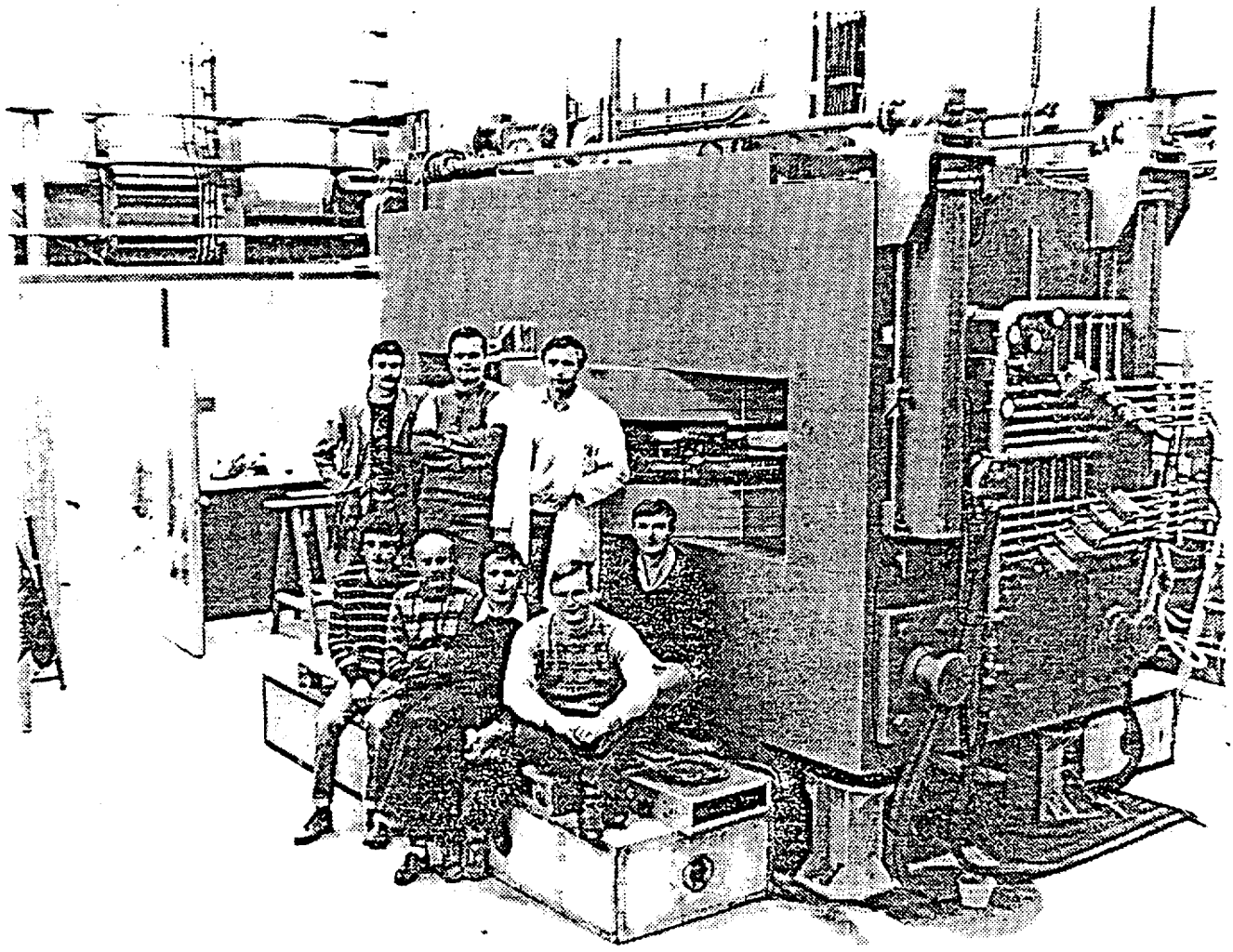
YEARS	1984	1985	1986	1987	1988	1989	1990
	STUDIES						
	-----	----	CONSTRUCTION				
COST IN		---	-----	----	-----	---	
1983 MFF	2.3	8.5	17.5	17.5	13	11.1	

The total cost is 69.9 MFF, based on 1983 prices. Including many uncertainties, a general cost of about 9 M US$ is foreseen excluding salaries of the personnel involved :

Engineers	: 17
Senior technicians	: 8
Technicians	: 20
Draughtmen	: 8
Craftmen	: 8

b) The cost of the MEDICYC cyclotron

The construction of MEDICYC has started in 1981 with a reduced team. Now the construction of the main components of the machine has been achieved (Figure 11). The two radiofrequency cavities have been



View of the cyclotron with the MEDICYC team

FIG. 11.

tested, the ejection system is partly manufactured and the magnetic measurements have been started. 10 persons have been working during 2 years on the construction:

1 Engineer

4 Senior technicians in mechanics

4 Senior technicians in electronics

1 Mechanic craftman

Two more years with more persons for mounting the remaining components of the machine are necessary. The following table summarizes the cyclotron costs :

MAGNET STEEL (120 TONES)		*1600.- R*
MAIN MAGNET COIL		*450.- R*
MAIN MAGNET POWER SUPPLY		*100.- R*
	TOTAL	*2150.-*
TRIM COILS (10)		*120.- R*
TRIM COILS POWER SUPPLIES		*400.- R*
	TOTAL	*520.-*
MAGNETIC MEASUREMENT SYSTEM		*220.- R*
	TOTAL	*220.-*
RADIOFREQUENCY SYSTEM		*1800.- R*
RF POWER SUPPLIES		*1200.- R*
LOW LEVEL SIGNAL CONTROL		*300.- R*
	TOTAL	*3300.-*
ELECTROSTATIC CHANNEL (ESC)		*220.- R*
ELECTROMAGNETIC CHANNEL (EMC)		*400.- R*
EMC POWER SUPPLY		*50.- R*
EXTRACTION SUPPORT		*115.- R*
INTERCONNECTING CABLES		*30.- R*
	TOTAL	*815.-*
INJECTION		*500.- P*
	TOTAL	*500.-*
VACUUM TANK		*300.- P*
PUMPS		*400.- P*
	TOTAL	*700.-*
HARD AND SOFT ELECTRONIC CONTROLS		*650.- R*
CONTROL DESK		*100.- R*
DIAGNOSTICS		*300.- P*
	TOTAL	*1050.-*
	TOTAL	*9255.-KFF*

R= Realized, P= Previsions

Nota: Salaries are not included, as usual in industry. This list concerns the cyclotron only.

V. CONCLUSIONS

The two cyclotrons presented in this paper are in fact quite different machines.

The nuclear physics field is always highly demanding. In a period of economic restraint it calls for high performing machines. The following table is a tentative comparison between a room temperature separated sector cyclotron and a cryogenic cyclotron.

CYCLOTRON TYPE	ROOM TEMPERATURE	CRYOGENIC
B AVERAGE (TESLA)	1.6	4 TO 5
TOTAL WEIGHT (TONS)	> 1200	~ 300
MACHINE DIAMETER (M)	~ 6	~ 2
ELECTRICAL POWER (MW)	> 1.5	~ 0.5

Cryogenic technology is in the advanced development stage but still requires specialised operating personnel, especially for the refrigerating and cryogenic networks. But the pionnering MSU I operates and the magnet of MSU II K 800 also. At Milan a K 800 cryogenic machine is in construction.

The above mentioned machines were designed to accelerate heavy ions. The Orsay design, which allows light ions beams with the axial injection in a unique central geometry and with its original magnetic field configuration (3 main coils, pseudo 3 sectors) represents a second generation of compact superconducting cyclotron and copes with the challenge of a relatively low cost (9 M US$).

For the medical field, a more classical solution has been chosen with a room temperature magnet. Nevertheless a new simple axial injection system has been used with a view to separate some of the main functions of the cyclotron and this should lead to a more reliable and performant machine with an overall cost of approximately 1.4 Million US dollars.

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PRODUCTION OF RADIOISOTOPES USING A CYCLOTRON

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Abstract

Cyclotron produced radioisotopes are generally neutron deficient and decay by EC or β^+ emission. They find major applications in diagnostic nuclear medicine. The production processes involve rather sophisticated technology and the areas needing research and development work include nuclear data, targetry, chemical processing, remote control, automation and quality control. A comparison of the various parameters relevant to the production of radioisotopes using a nuclear reactor and a cyclotron is given. The cyclotron products are more expensive than the reactor products; they are, however, far superior to the latter as far as in-vivo functional studies are concerned.

INTRODUCTION

Cyclotron produced radioisotopes are generally neutron deficient and decay by electron capture or β^+ emission. If they meet the general criteria [cf. 1] for medical applications, i.e. short half-life; E_γ between 60 and 500 keV or β^+ emission [for detection with a γ-camera or emission tomography: single photon emission tomography - SPECT - for single photons in the energy range of 100-200 keV (^{99m}Tc, ^{123}I) and positron emission tomography - PET - in the case of positrons]; suitable biological properties, they are of great significance for in-vivo metabolic studies. Although some long-lived cyclotron products like $^{22}Na(T_{1/2}$ = 2.6 y), $^{54}Mn(T_{1/2}$ = 312 d), $^{55}Fe(T_{1/2}$ = 2.7 y), $^{57}Co(T_{1/2}$ = 270 d) etc. also find technological applications, the major use of cyclotron radioisotopes is in diagnostic nuclear medicine.

All the radioisotopes used for in-vivo studies can be arbitrarily divided [cf. 2] into five groups according to their chemical behaviour, biological function or mode of formation (cf. Table I). The "organic" short-lived β^+ emitters (e.g. ^{11}C, ^{15}O etc.) are ideally suited for labelling biomolecules and find

applications in PET. Radiohalogens may also be regarded as "organic" isotopes since they are also useful for labelling biomolecules; some of them (e.g. ^{123}I) are suitable for in-vivo studies using conventional γ-cameras or SPECT while the others (e.g. ^{18}F, ^{75}Br) find applications in PET. Rare gases are generally used for ventilation studies. Since many of the radiohalogens are formed via rare gas precursors, these two types of radioisotopes are grouped together. Short-lived generator radionuclides are practical for general medical use since an on-site or nearby cyclotron is not required. Alkali and alkali like metals find applications in myocardial perfusion studies. The list of inorganic radionuclides is large but their applications are rather limited.

TABLE I

Types of Cyclotron Produced Radionuclides for in-vivo Studies

1. "Organic" short-lived β^+ emitters	(^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{30}P etc.)
2. Halogens and rare gases	(^{18}F, ^{34m}Cl, $^{75,77}Br$, ^{123}I, $^{77,79}Kr$, ^{123}Xe etc.)
3. Generator isotopes	(^{68}Ge-^{68}Ga, ^{81}Rb-^{81m}Kr etc.)
4. Alkali and alkali like metals	($^{38,43}K$, ^{81}Rb, $^{128,129}Cs$, ^{201}Tl etc.)
5. "Inorganic" radionuclides	(^{28}Mg, ^{45}Ti, ^{48}Cr, ^{67}Ga, ^{73}Se, ^{97}Ru, ^{117m}Sn etc.)

The short-lived organic β^+ emitters can be produced using small cyclotrons ($E_d \leq 10$ MeV, $E_p \leq 16$ MeV); for the others, however, higher energy cyclotrons are needed.

PRODUCTION

The production of radioisotopes using a cyclotron involves rather sophisticated technology and for reaching the stage of routine production considerable research and development work in the following areas may be needed.

Nuclear Data

A radioisotope may be formed via several nuclear reactions, their number increasing with the increasing energies of the available accelerated particles. Not every reaction, however, is suitable for routine production. In practice one is generally limited to the use of one or more of the four projectiles p, d, ^{3}He and ^{4}He. From the viewpoint of nuclear data the important criteria for choosing a particular process consist of high yield of the desired radioisotope and minimum possible contamination from the accompanying impurities. Whereas the non-isotopic impurities produced can be removed by chemical separations, the level of isotopic impurities can be suppressed generally by using enriched isotopes as target materials and/or by a careful selection of the particle energy range effective at the target. Evidently it is necessary to know the excitation functions of the various competing reactions accurately (for a detailed discussion on nuclear data cf. [1,2]).

The production of ^{123}I($T_{1/2}$ = 13.2 h) via the $^{124}Te(p,2n)^{123}I$ reaction, using highly enriched ^{124}Te, furnishes a good example of the importance of nuclear data. Due to the competing $^{124}Te(p,n)^{124}I$ reaction it is not possible to eliminate the ^{124}I-impurity($T_{1/2}$ = 4.15 d) completely, even if ^{124}Te is 100% enriched. Fig. 1, based

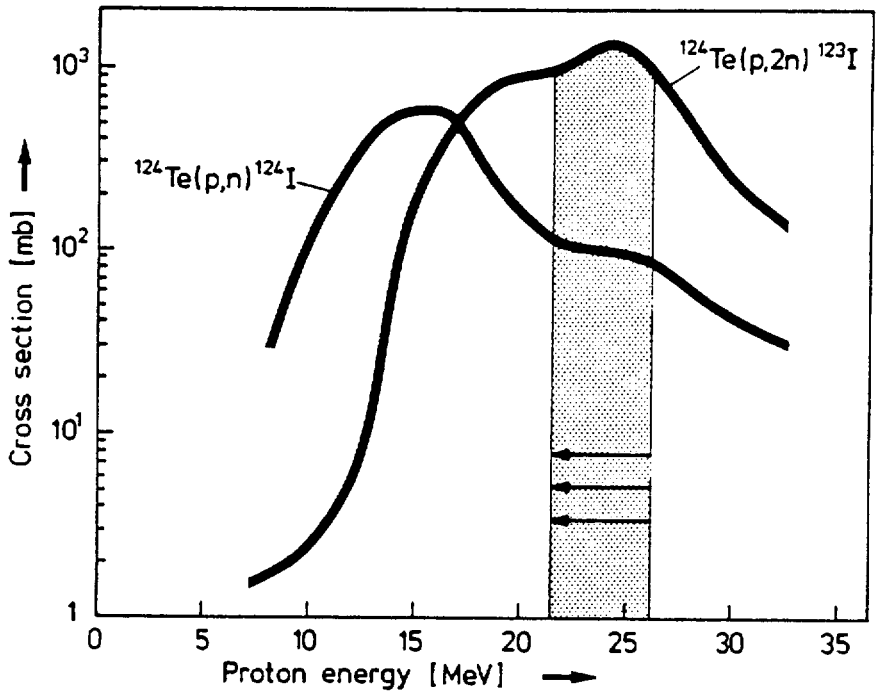

Fig. 1 Excitation functions of $^{124}Te(p,n)^{124}I$ and $^{124}Te(p,2n)^{123}I$ reactions. The optimum energy range for the production of ^{123}I is E_p = 26.2 → 21.5 MeV [3].

on the excitation function measurements of Kondo et al [3], shows that the ideal proton energy range for the production of ^{123}I is E_p = 26.2 → 21.5 MeV, i.e. the energy of the incident protons should be selected as 26.2 MeV and the thickness of the tellurium target should degrade the incident energy only to 21.5 MeV. For a 99% enrichment of the target the level of undesired ^{124}I-impurity in this energy range is expected to be ∿ 0.5%.

Targetry

Design and construction of a target capable of withstanding high beam currents is one of the most crucial steps in the large scale production of radioisotopes. Several nuclear processes, though very promising from the viewpoint of yield and impurity level, may not be used if target technology is not sufficiently developed. If the target material consists of a high melting metal, construction of a suitable target may not cause great difficulty. Similarly if the energy of the available projectile is sufficiently high, 4 π water cooling may be introduced so that an efficient heat dissipation is achieved. In many cases, however, some or all of the following points may need careful consideration.

- Energy of the projectile. If it is low, only a thin window may be allowed between the cyclotron vacuum and the target material. The window may be ruptured occasionally.
- Pressure within the gas target. If it is high, there may be need for a double window system, cooled by He flow.
- Melting point of the target metal, and thermal conductivity if in oxide form.
- Cost of the highly enriched material. Expensive targets demand minimum loss during irradiation and subsequent chemical processing, as well as efficient regeneration of the target.
- Half-life of the product. A short half-life requires an efficient and fast chemical separation.
- Chemical reactivity of the products.

We give below examples of some typical target systems to show the nature of the problems involved.

Production of short-lived β^+ emitting gases like ^{11}C, ^{15}O etc. is done on-line in a flow system [cf. 4]. The arrangement for ^{11}C production via the $^{14}N(p,\alpha)^{11}C$ reaction is shown in Fig. 2.

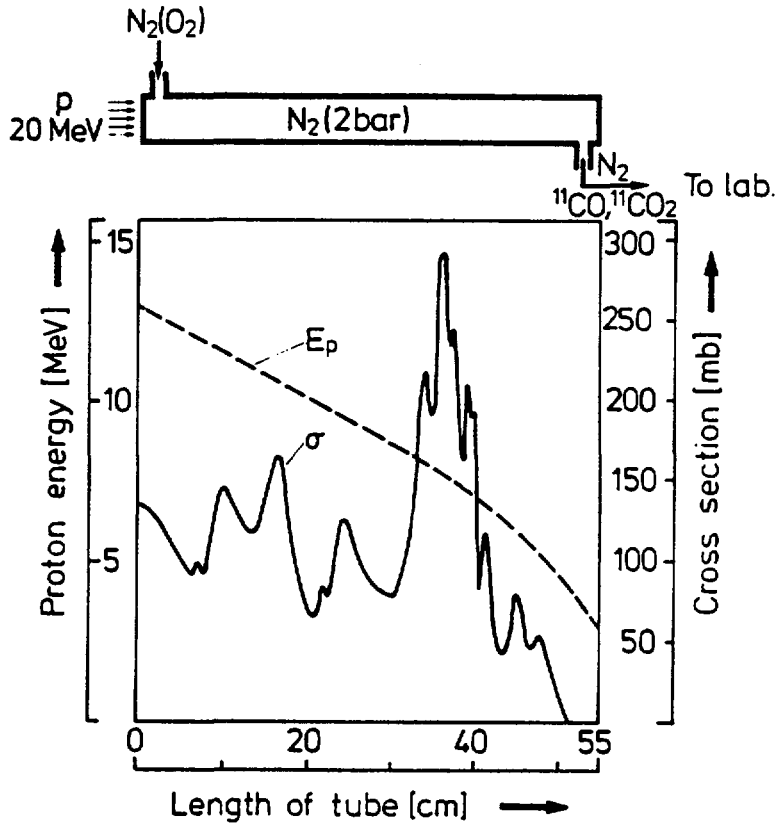

Fig. 2 Schematic diagram of a gas target used for the production of ^{11}C via the $^{14}N(p,\alpha)^{11}C$ reaction. The cross section [6] and the absorption of the protons in the target are shown as a function of the target length.

The target consists of a 50 cm long tube through which N_2 gas (containing traces of O_2) flows at a pressure of 2 bars. A 30 μA beam of 20 MeV protons passes through a thick window and is incident on the gas target with an energy of 13 MeV. The length of N_2-filled target corresponds to E_p = 13 → 4 MeV and covers the whole excitation function. ^{11}C is produced as ^{11}CO + $^{11}CO_2$ and is carried through a capillary tube to the laboratory area where chemical synthesis is performed, generally after conversion to $^{11}CO_2$ or other precursors [cf. 5].

Ne gas targets are commonly used for the production of ^{18}F via the $^{20}Ne(d,\alpha)^{18}F$ reaction [for a review cf. 7]. In this case the target generally consists of a tube filled with about 18 bar Ne. A double window (with He cooling, 6 bar) is used between the Ne gas and the cyclotron vacuum. The radiofluorine is very reactive and, in the absence of any carrier, sticks to the inner walls of the target, from where it is rather difficult to remove. The inner

walls of the target therefore need special treatment, for example, passivation with fluorine, lining with nickel etc. Use of F_2 carrier in the range of 0.1 to 0.75% leads to the removal of ^{18}F-labelled F_2 [cf. 8,9].

Liquid targets are not common, mainly due to hydrolysis and radiation chemical effects. Production of ^{18}F via $^{16}O(^3He,p)^{18}F$ and $^{18}O(p,n)^{18}F$ reactions using water targets has, nonetheless, been performed. The latter target is of special interest for producing ^{18}F at a small cyclotron. These targets are, however, of limited use for further synthetic application of ^{18}F, since the solvated fluoride is very unreactive and the water has to be removed completely. Molten salt target systems have been used occasionally for the production of radiobromine and radioiodine. In such systems, however, special corrosion resistant materials have to be used.

For production of radioisotopes other than short-lived β^+ emitters solid target materials are commonly used. Fig. 3 gives a sketch of the target system [cf. 10] used for the production of ^{123}I via the $^{124}Te(p,2n)^{123}I$ process. The target material $^{124}TeO_2$ (with ^{124}Te enrichment of 99.9%) is molten on a Pt backing and adheres to it well. The front side of the target is in direct contact with a layer of flowing cooling water. The back side is also cooled by water. In the case of a compact cyclotron, having the maximum proton energy lower than that ideally required for the production process, the front water layer has to be rather thin, otherwise the protons would lose too much energy

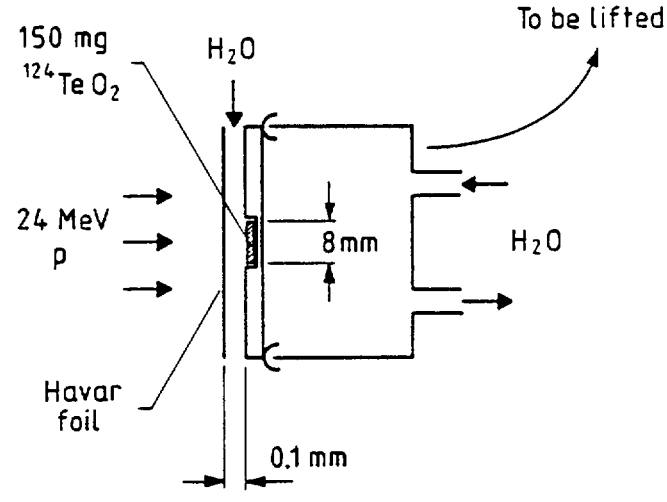

Fig. 3 Simplified sketch of remotely controlled high-current target system used for irradiations of $^{124}TeO_2$ [10].

before reaching the surface of the target material. The target system can withstand high beam currents: the effective power density for a 24 MeV proton beam of 30 μA amounts to ∿ 1400 W cm^{-2}. The loss of $^{124}TeO_2$ during a 1 h irradiation at 30 μA is $\leq$ 1%.

In order to be able to use low melting and poor thermal conducting elements as target materials, two approaches have been developed recently. In the first, the extracted beam is defocussed, wobbled and allowed to fall slanting on a large sized target, which is cooled efficiently at the back and rotated. An example is the production of high-purity ^{77}Kr via the $^{77}Se(^3He,3n)^{77}Kr$ reaction [11]. The target consists of a thin layer of metallic ^{77}Se (94.4% enriched) on an Al backing and the ^3He-beam falls at an angle of 19°. For beam currents up to 20 μA the loss of ^{77}Se is $\leq$ 1%. In the other approach, the element is transformed into a suitable alloy and irradiated in the usual way. The production of ^{75}Br via the $^{75}As(^3He,3n)^{75}Br$ reaction serves as a good example [cf. 12]. A high melting Cu_3As-alloy (with the optimum As-content of ∿ 31%) was developed. A thin layer of this alloy, covering the optimum energy range of $E_{^3He}$ = 36 → 25 MeV, is prepared on a Cu-backing by melting in a hydrogen flame. The target is capable of withstanding high beam currents: the effective power density for a 36 MeV ^3He-particle beam of 35 μA amounts to ∿ 2100 W cm^{-2}.

It should be mentioned that in many production processes use is also made of internal beams which are of much higher intensity than the extracted beams. Fig. 4 shows, as an example, the internal irradiation system used for the production of ^{75}Br [cf. 13]. The target consists of a thin layer of Cu_3As-alloy on a wedged Cu-backing and the ^3He-particle beam falls at an angle of 6.2°. Irradiations are performed routinely at 100 μA; beam adjustment is done using a thermoelement.

The above discussion should demonstrate the complexity of problems associated with the construction of high-current targets. Thus, considerable experience, skill and development work are essential prior to embarking on the production of radioisotopes using a cyclotron.

Chemical Processing

The short-lived β^+ emitting gases (^{11}CO, $^{11}CO_2$, ^{15}OO, $^{13}NH_3$) are generally separated by on-line removal from the gas targets [4]. The flow rate of the gas through the capillary tube, the

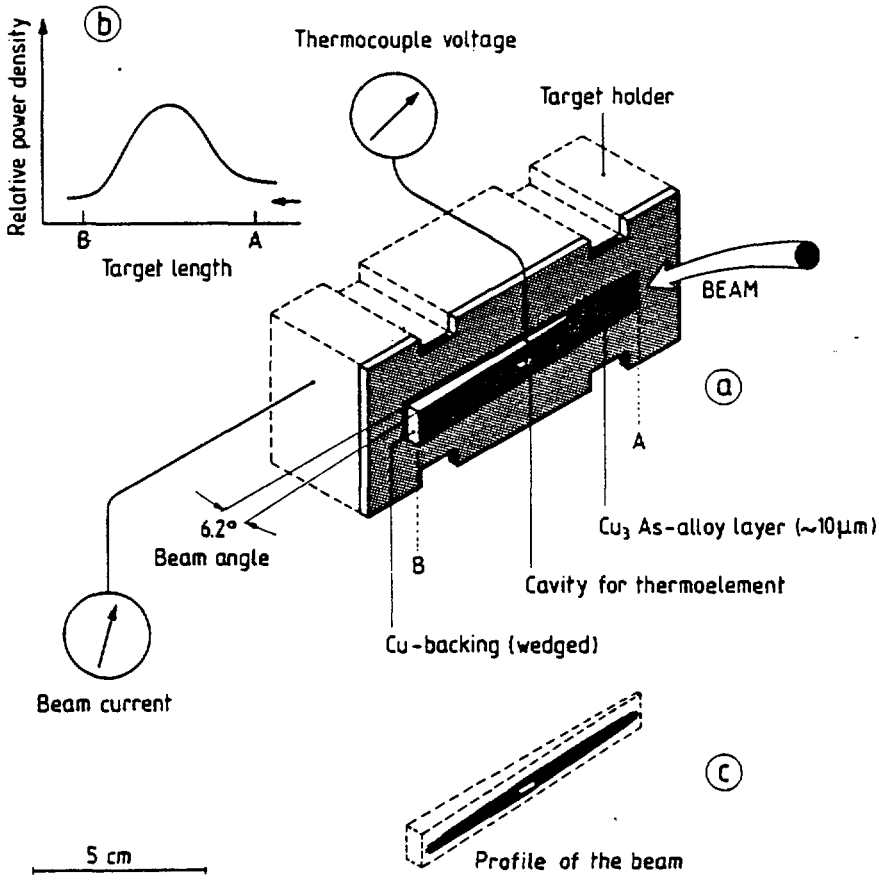

Fig. 4 a) Sketch of target and target holder used for irradiations with internal beams. b) Relative power density experienced by the thermoelement. c) Autoradiographic profile of the beam incident on the target [13].

use of filters and cold traps, and the shielding of the system are some essential considerations.

On-line removal of gaseous products from solid or molten targets has also been used. The prduction of ^{123}I via the ^{123}Xe-precursor, for example, involves the removal of radioxenon from a NaI or KI target irradiated with high energy protons or deuterons [cf. 14,15]. In one modification a solution of CH_2I_2/I_2 or LiI/I_2 flowing in a loop is used as target material [cf. 16]. A stream of He gas passes through the target material and carries radioxenon with it, which is subsequently adsorbed in a liquid nitrogen trap. After a suitable decay period, ^{123}I is removed from the trap.

Separation of a radioisotope in a batch process is also quite common. Due to the high level of radioactivity involved, a one step separation procedure is always preferred. In recent years, the dry distillation technique has been gaining considerable significance. The separation of "no-carrier-added" ^{75}Br from the

irradiated Cu_3As-alloy, for example, is performed via distillation at 950 °C [12]. Radiobromine separated from the target is carried over by a helium stream and condenses in a quartz tube from where it is taken up in a small volume of water. The radiochemical yield of this thermochromatographic method is > 90%. The distillation method is of great advantage when the target material is isotopically enriched since, after the separation of the radioisotope, the same target can be reused for subsequent production runs. The separation of ^{123}I from the irradiated $^{124}TeO_2$ [cf. 10,17] furnishes a good example. A sketch of the apparatus used is shown in Fig. 5. The loss of $^{124}TeO_2$ during distillation is < 1% and the radiochemical yield is > 95%.

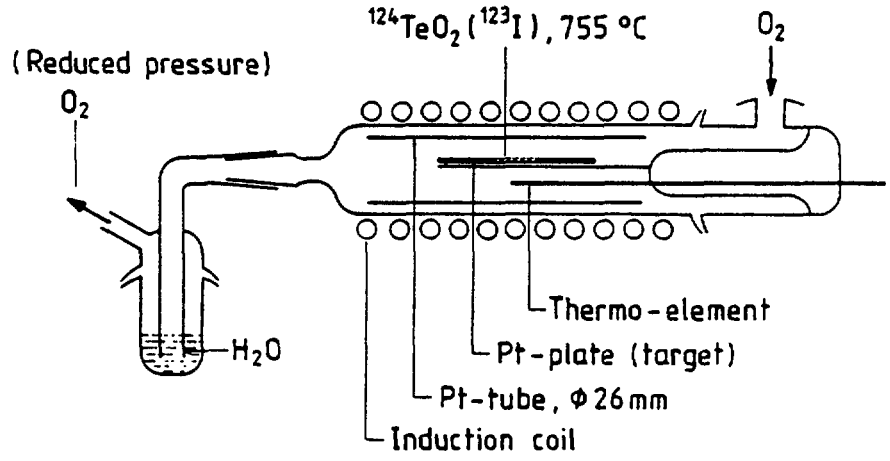

Fig. 5 Sketch of quartz apparatus used for dry distillation of ^{123}I from irradiated $^{124}TeO_2$ [cf. 10,17].

Often several step wet radiochemical methods of separation are unavoidable, especially when the target and the product have very similar vapour pressures, or when the energy of the projectiles used is high, so that a large number of radioactive products are formed. The production of ^{28}Mg, ^{48}Cr and ^{201}Tl via medium energy reactions and of ^{77}Br in the spallation process constitute typical examples. Several separation techniques like coprecipitation, ion-exchange chromatography, solvent extraction, extraction chromatography, high-pressure liquid chromatography etc. are applied and in some cases the methods are quite elaborate. Since separations are done at "no-carrier-added" level, the final radiochemical yields generally vary between 30 and 90%.

Remote Control and Automation

As mentioned above, the desired radioisotopes are generally short-lived. In order to be able to achieve sufficient quantity of the final product (i.e. after the chemical separation and labelling steps, during which the radioisotope is decaying) it is necessary to have rather high initial activities. In the production of ^{11}C-labelled compounds, for example, it is not uncommon to have initial activity on the order of 1 Ci. Similarly, the dose rates on the surface of solid irradiated samples are often > 1000 R/h. In the case of β^+ emitters 511 keV annihilation radiation is present. In short, the level of radioactivity is rather high. Radiation protection regulations demand the use of remotely controlled methods. Radiochemical work has to be done in lead cells.

Since during both irradiation and subsequent chemical processing a large number of operations and checks are involved, use of small computers for automation of the production process is highly advantageous.

Quality Control

In general three types of quality control measures are necessary.

TABLE II

Radionuclidic Composition of Radioiodine Produced via commonly used Processes

Reaction	Target enrichment (%)	Radionuclidic content (%)*					
		^{123}I	^{124}I	^{125}I	^{126}I	^{130}I	^{131}I
Indirect							
$^{127}I(p,5n)$	Natural	99.3	-	0.7	-	-	-
$^{127}I(d,6n)$	Natural	98.8	-	1.2	-	-	-
Cs,La(p,spall)	Natural	98.8	-	1.2	-	-	-
Direct							
$^{124}Te(p,2n)$	96.5	96.2	3.8	-	-	-	-
$^{123}Te(p,n)$	91.5	98.3	1.7	-	?	?	?
$^{122}Te(d,n)$	96.5	97.5	0.4	-	0.3	1.4	0.4

*30 hours after bombardment, optimum particle energy

a) Radionuclidic purity. This is generally done using high-resolution γ-ray spectroscopy. Occasionally the level of radionuclidic impurity influences the decision whether a radioisotope should be used at all. If a certain impurity associated with a particular production method causes high radiation dose or affects the quality of the scans, the very advantage of the short-lived radioisotope may be jeopardized. In such a case an alternative method of production has to be looked for. Table II gives, as an example, the impurities present in ^{123}I produced via various nuclear processes. The indirect method of production, i.e. via the $^{123}Xe \xrightarrow[2.1\ h]{\beta^+,EC} {}^{123}I$ process, is preferred since the only associated impurity ^{125}I does not cause much radiation dose to the patient, when present at ∿ 0.2% at EOB.

b) Radiochemical purity. The separated radioisotope must be in a well defined radiochemical form, otherwise difficulties may arise in the subsequent labelling work. Some oxidation/reduction cycle may be necessary to achieve this at the "no-carrier-added" level. Several radiochromatographic techniques are used to determine the radiochemical form of the species present. It has been found, for example, that ^{75}Br taken up in water after the thermochromatographic separation exists > 95% as bromide.

c) Chemical purity. In addition to radionuclidic impurities, the separated radioisotope may contain some inactive isotopic and non-isotopic impurities. A careful check of those impurities is necessary. In this connection both nuclear and non-nuclear methods of analysis are needed. The most common methods include activation analysis, spectrophotometry, atomic absorption spectroscopy, optical emission spectroscopy, etc. Special attention has to be paid to detecting toxic substances present in the final products.

The undesired inactive isotopic impurities decrease the specific activity of the product. Strictly carrier-free products are hard to obtain. It is, however, very important that the products are at the "no-carrier-added" level.

In the case of labelled compounds prepared for applications in humans, sterility and apyrogenecity are also mandatory.

COMPARISON OF CYCLOTRON AND REACTOR PRODUCTION OF RADIOISOTOPES

A comparison of various parameters relevant to the production of radioisotopes using a cyclotron and a nuclear reactor is given in Table III. In general it may be said that production using a

TABLE III

Comparison of Cyclotron and Reactor Produced Radioisotopes

Parameter	Cyclotron Radioisotopes	Reactor Radioisotopes
A. General		
Decay mode	EC, β^+, IT	β^-, IT
Half-life	short	long (except generator systems)
Radiation dose	small	variable (generally high)
B. Production		
Nuclear data	extensive charged particle data base required	(n,γ), (n,p), (n,fission) processes; data available
Targetry	sophisticated technology	rather easy
Chemical processing	on-line or fast separation essential; in spallation process elaborate separation schemes	conventional methods sufficient; in fission process very elaborate separation schemes
Remote control	essential	sometimes essential
Shielding	β,γ shielding essential	β,γ shielding essential; in fission process also α shielding
Quality control	radionuclidic, radiochemical and chemical purity checks	radionuclidic, radiochemical and chemical purity checks; in fission process control of α-emitters
Specific activity	generally high	generally low (except generator systems and fission products)
Cost	generally high	relatively low
C. Application		
Areas of use	primarily in-vivo diagnostic nuclear medicine	in-vitro methods (clinical chemistry, biochemistry, pharmacology) nuclear medicine (diagnosis and therapy); technology and industry
Detection	in-vivo γ-scanning; SPECT; PET	in-vivo γ-scanning; SPECT in some cases; in-vitro β^- counting
Functional studies	short-lived β^+ emitters and halogens useful for in-vivo metabolic studies	long-lived β^- emitting ^3H and ^{14}C useful only for in-vitro investigations
D. Distribution		
Availability	occasional machine breakdown possible, production affected	production more reliable
Logistics	fast transport essential	longer half-life makes transport easier
Waste disposal	no problem	special methods essential

cyclotron involves more sophisticated technology and more expense. The products, on the other hand, are far superior to the reactor products, as far as in-vivo applications in nuclear medicine are concerned.

The absence of any suitable γ-ray emitting reactor radioisotope of organic elements leaves the choice for in-vivo investigations in man only on ^{99m}Tc ($T_{1/2}$ = 6.0 h) and - for therapy mainly - ^{131}I ($T_{1/2}$ = 8.05 d). The former isotope is, however, not a good

label for organic compounds, and hence not very suitable for functional imaging, but still finds wide application. On the other hand, the β^- emitters $^{3}H(T_{1/2} = 12.4\ y)$ and $^{14}C(T_{1/2} = 5736\ y)$ are still very important reactor radioisotopes for in-vitro studies in biochemistry, pharmacology, etc. Of special importance is the use of $^{125}I(T_{1/2} = 60.1\ d)$ in radioimmunoassay, one of the most important clinical in-vitro methods of very wide application.

The cyclotron radioisotopes like ^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{75}Br and ^{123}I are not only good labels for organic compounds but also cause low radiation dose. Furthermore, in the case of β^+ emitters and suitable photon emitters, emission tomography leading to three dimensional information can be applied.

It needs also to be pointed out that the short half-lives of cyclotron products cause considerable logistics problems. The β^+ emitters ^{11}C, ^{13}N and ^{15}O can be used only on the site of production; ^{18}F and ^{75}Br could be transported to hospitals in the vicinity of about 100 km. For more general applications of cyclotron radioisotopes, however, presently the choice lies on ^{67}Ga, ^{123}I and ^{201}Tl, or on generator systems like $^{68}Ge/^{68}Ga$, $^{81}Rb/^{81m}Kr$ etc. Due to these mundane limitations on the one hand, and the many in-vitro methods used in clinical chemistry and biochemistry on the other, it seems that in the immediate future both reactors and cyclotrons will continue to compete as sources of medical radioisotopes applicable on a more general basis.

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D-T NEUTRON SOURCES AS AN ALTERNATIVE TO RESEARCH REACTORS

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Abstract

The D-T neutron generators used by small resarch laboratories are characterized by the following performances. Accelerating voltage: 150 - 400 kV, beam current ≤2mA useful long term neutron yield -10^{11} in 4π sr. The discussion of the potential of neutron generators for research and application is limited to the scope of machine parameters discussed above.

The following topics are discussed:

- **nuclear data measurements, microscopic and macroscopic data,**
- **uses as analytical tool in geology, environment, agriculture and other fields where they can compete effectively with other analytical techniques,**
- **uses as radiation sources for studies in radiobiology and medical applications,**
- **miscellaneous.**

The discussion includes the aspects of cost, maintenance skills required, existence of regional activities, etc.

The review of the areas of work with neutron generators shall be restricted to those usually found in small laboratories, i.e. accelerators with terminal voltage <400 kV, most usually 150-200 kV, with deuteron currents of up to 1 mA. Additionally, most of those generators are not equipped with pulsing facilities and a magnetic analysis of the beam is seldom available.

Of course it is difficult to assess the scope of research one can do with a given research tool. The use shall depend probably more on the environment, the current interest and profiles of the people gravitating around the tool. An active medical, biology, geology etc. department shall necessarily influence the research in a given direction.

Such a view stems from the belief that the intrinsic potential of such accelerators for work in the field of nuclear physics is too small to warrant their exclusive use in the narrow field of nuclear physics. This is of course also true for the research reactors.

We shall try to illustrate the potentialities of the use of fast neutrons in the following fields:

- element analysis;
- nuclear data and nuclear reactions;
- radiation damage in electronic components;
- radiobiology and dosimetry;
- benchmark experiments;
- miscellaneous.

Elemental analysis

Elemental analysis is of fundamental importance in many multidisciplinary research projects as well as applications. The nuclear technique have brought to elemental analysis three powerful methods; X-ray fluorescence (XRF) induced by photons, thermal neutron activation (TNA) and fast neutron activation (FNA). All three techniques are applicable to a wide range of elements but in general one may say that the XRF shall be the simplest to apply for Z>15, that the TNA is applicable accross the periodic system except for specific cases, which fortunately are amenable to activation by fast neutrons. The fast neutrons can also be used in a fairly wide range but one must bear in mind that the achievable fluences and the relevant cross sections are very much smaller for fast neutrons than for

thermal neutrons so that the limits of detection in FNA are inferior to those in both TNA and XRF.

We shall dwell here only with those elements where the FNA has a definite edge over the other two techniques.

It should be specially borne in mind that, for daughter nuclei, having not a too short half life, sending a limited number of samples to a reactor for irradiation, followed by counting in the home lab, may still be preferable compared with the inadequate use of a neutron generator, since the thermal neutron fluxes achievable with a neutron generator equipped wiht a moderating assembly is at best two orders of magnitude smaller than the fast neutron fluxes. An extensive review of the literature on activation analysis with fast neutrons is given in ref. 1.

There is a high potential for the use of elemental analysis in many domains of industrial production that are within the reach and interest of developing countries. There exist operative systems for nitrogen analysis by way of checking the protein content of foods. The metallurgical companies use extensively the neutron activation for oxygen determination in iron. We shall review here some of the elements which are most amenable to FNA. Our review shall stop at A=30 because beyond that unless very specific needs appear the relatively cheap and simple XRF shall supplant the results that can be obtained by fast neutrons. Compared to TNA, fast neutron analysis is advantageous for elements like niobium, thallium etc. (cf. ref. 2) for which the (n,γ) process leads to β^- emitters. Furthermore, in the region of rare earths (n,γ) processes often yield radioisotopes with similar half-lives and γ-rays. The use of the $(n,2n)$ process in FNA may lead to higher precision.

Carbon

The analysis of carbon is the most complicated since it uses the detection of the prompt gamma ray emitted in the

process $^{12}C(n,n'\gamma)$. The energy of the gamma ray that is detected is 4.43 MeV.

Cleaner data can be obtained with the use of pulsed neutron generators. Using large samples (several tens of kilograms) one may detect concentration of tens of percents using a NaI crystal positioned to view the sample that is in beam.

The method has been used to measure the carbon content of coals[3].

Nitrogen

Nitrogen built in the proteins is one of the most important elements in our food chain. The fertilizers, the plant and animal breeding are geared towards obtaining a large protein content in our food. The correlation that exists between nitrogen and the protein content makes the analysis of the former very necessary. There are many nuclear techniques for nitrogen analysis[4] but the $^{14}N(n,2n)^{13}N$ reaction with fast neutrons is by far the best investigated and most widely used activation analysis method[5].

Although relatively simple in putting to work, ^{13}N is a β^+ emitter with 10 min half life and one has to reckon in practice with several interfering reactions that interfere either by β^+ emitters of similar half life or by resulting also in ^{13}N activity. These reactions are $^{31}P(n,2n)^{30}P$, $^{39}K(n,2n)^{38}K$, $^{13}C(p,n)^{13}N$, $^{12}C(p,\gamma)^{13}N$ and $^{63}Cu(n,2n)^{62}Cu$. Especially disturbing are the proton induced reactions caused by recoil protons in carbohydrate matrices or in tissues containing water, where large amounts of H, C and O are present.

Another possible interference is from the activation of the air nitrogen present in the container. For all these reasons the determination of nitrogen is generally limited to concentrations of ~0.5%[6].

Oxygen

The determination of oxygen has a vast field of application. The field where it is most commonly used is the determination of oxygen in iron, but we should like to point out an application that seems useful for geological studies and feasible in a non industrial environment. Namely, from about 2000 kinds of minerals in the earth crust at least 1200 contain oxygen. In spite of its high importance the knowledge about the contant of oxygen is rather poor because it is very difficult to measure the oxygen concentrations in rocks by chemical analysis. In most cases the oxygen content is calculated from the oxide measurements with a fairly large error[7].

The reaction $^{16}O(n,p)^{16}N$ reaction is used for the determination of oxygen. The high energy gamma rays of 6.13 MeV following the β decay of the ^{16}N nuclei can be detected without any interference from other elements except for the fluorine which may give the same residual nucleus via the reaction $^{19}F(n,\alpha)^{16}N$. However, if this is the case the amount of fluorine can be independently determined via the (n,p) and/or (n,2n) reactions.

Because of the short half life of $^{16}N(\tau_{1/2}=7.4$ s) the oxygen determination necessitates the use of the so called cyclic activation[8] which means that a rapid (~1 s) rabbit system should be available so that the same sample may be irradiated and counted several times (typically ~10 times) until sufficient statistics is accumulated. Due to the high energy of the gamma rays it is necessary to use NaI detectors and not Ge.

Fluorine

Fluorine is found in many compounds of the modern technology and also believed to be important from the aspect of dental health. The reactions that are available for the de-

termination of fluorine are the following $^{19}F(n,p)^{19}O$, $^{19}F(n,\alpha)^{16}N$ and $^{19}F(n,2n)^{18}F$[9]. The first two reactions cannot be used without a rabbit system because of the short half life, 30 s and 7.4 s, respectively.

Magnesium

Magnesium is an element often met in nature and vital for life. Also in the metallurgical industry the basic parameters of the spherical cast iron depend on the magnesium concentration[10].

Magnesium analysis is easily achievable via the $^{24}Mg(n,p)^{24}Na^{g}$ reaction.

Aluminium

The reactions $^{27}Al(n,p)^{27}Mg$ and $^{27}Al(n,\alpha)^{24}Na^{g}$ are suitable for aluminium determination using a standard Ge gamma ray detector. Quantities as low as ~20 μg have been measured during measurements of aerosol concentration[11]. The reaction $^{30}Si(n,\alpha)^{27}Mg$ may interfere with the results of the (n,p) reaction.

Silicon

Silicon is determined by $^{28}Si(n,p)^{28}Al$, $^{29}Si(n,p)^{29}Al$ and $^{30}Si(n,\alpha)^{27}Mg$ reactions[11]. The limit of detection is rather low ~10 μg[9].

The interfering reactions are the following: $^{27}Al(n,\gamma)^{28}Al$ and $^{31}P(n,\alpha)^{28}Al$. However, the variety of reactions make an unambiguous determination possible.

The determination of Si and Al finds easy applications in geological studies and aerosol studies.

Phosphor

The importance of phosphates for agriculture makes the search for adequate phosphate mining sites an imperative for many countries.

The reactions $^{31}P(n,p)^{31}Si$ and $^{31}P(n,\alpha)^{28}Al$ are available: the first one suffering from the interference of the $^{34}S(n,\alpha)^{31}Si$ reaction and the second from the interference of the $^{28}Si(n,p)^{28}Al$ reaction. The $^{31}P(n,2n)^{30}P$ is free from interference but results in a β^+ emitter.

Sulphur

Laboratories equipped with facilities for β^- counting shall find a lot of interesting studies in the determination of sulphur. Sulphur is determined by the $^{32}S(n,p)^{32}P$ or the $^{34}S(n,p)^{34}P$ reaction[12].

Nuclear data and nuclear reactions

A very high percentage of the time spent on neutron generators is devoted to the study of neutron induced reactions, (the spectra of outgoing particles, cross sections and their dependence on incident energy, spectroscopy). A large amount of that work is gnerated by the need for precise nuclear data for shielding, energy and dosimetric application. Part of the effort is on the other hand devoted to the study of the basic nuclear physics undelying the observed parameters. A detailed and exact description of neutron induced processes should reduce the tedious experimental collection of nuclear data. The work in this field can be divided in several broad topics.

- Measurement of neutron elastic and inelastic scattering cross section.
- Measurement of integrated cross sections for processes amenable to measurement by activation analysis.
- Measurement of (n, charged particle) and (n,gamma) reactions by the direct detection of emitted partices or γ-rays.

A rather good overview of the latest work done in these areas is given in the proceedings of the International Conference "Nuclear Data for Science and Technology"[13] and refs. 14, 15, 16 and 17.

I shall dwell here on only a couple of aspects that are worthwile to be considered as relatively new developments in the field and could be of interest to laboratories looking for new directions in their work with fast neutrons.

Measurement of gamma spectra and gamma multiplicities in (n,n'xγ) reactions

It has been recently demonstrated[18,19] that it is possible to measure the multiplicity of gamma rays emitted in a (n,n'γ) reaction using a relatively simple setup. The setup is shown in Fig. 1 and consists of a neutron detector, a Ge detector and NaI detector. The time of flight measurements for the neutrons are done using the associated particle method for timing. The Ge detector is used to identify a given transition

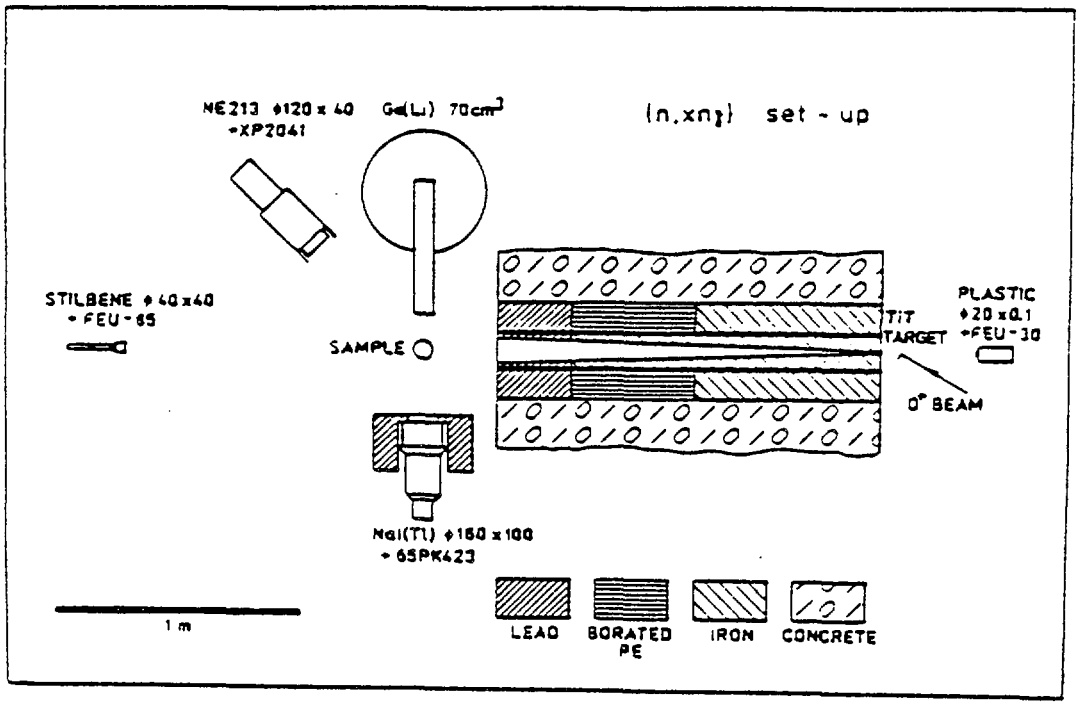

Fig. 1. Experimental arrangement.

in the residual nucleus and the NaI detectors the continuum gamma rays. With two sets of coincidence requirements the authors have shown that it is possible to obtain new and interesting information on the multiplicity of the emitted gammas.

Measurement of characteristic x-rays emitted after neutron activation

There are cases when the activated sample does not emit gamma rays as is the case for electron capture processes in some cases. Only characteristic x-rays shall be detected. Also in other cases the detection of x-rays as a complementary datum to the one yielded by gammas may be fruitful[20].

The advent of high resolution x-ray detectors has made this work possible and desirable.

Study of low-yield reactions

In recent years low-yield reactions like (n,t), (n,^{3}He), (n,n'p), (n,n'α) etc. have been investigated extensively by the radiochemical method[16,21,22]. There is, however, still considerable scope for further studies.

Radiation damage in electronic components

The radiation effects of gamma rays and reactor neutrons on various novel electronic components have been fairly extensively studied. However, the same is not true for 14 MeV neutrons, probably, due to the general belief that very high fluences are necessary to induce damages. It has been lately demonstrated that for instance infrared emitting diodes and optocouplers have exhibited radiation damage in the range of neutron fluences of $10^{10} \div 10^{12}$ cm^{-2}[23]. Every day we are faced with new electronic components and the study of radiation damages with fast neutron may give insight in the mechanism of the damage and may in some cases enlarge the list of potential dosemeters.

Radiobiology and dosimetry

The relative biological efficiency is defined as the quotient of the biological effect of a given dose of neutrons and gamma rays. In general the RBE for neutrons is higher than unity, but its magnitude depends on the effect that is investigated (cell survival, chromosomic aberrations etc.), the dose administered, the dose rate, the type of tissue or cell culture investigated. The state of experimental data is not yet on the level that would make possible theoretical explanation that would go much beyond phenomenological interpretations. The field of radiobiology with fast neutrons is surely not exhausted and it is probable that a better understanding of the radiation effects and of the reparation mechanisms may come from such investigation especially in the field of low and intermediate doses.

The conditio sine qua non for a radiobiological investigation is the existence of a reliable dosimetry, the precision of the dosimetry should be $\pm 5\%$. Preferably one should use two types of independent dosimetric measurements (two ionization chambers, tissue equivalent calorimeter[24], or others[25]).

The requirements on the dose rate are rather high, but a satisfactory arrangement was obtained with a 800 μA beam on a rotating target at 11 cm from the target. Neutron rates achieved were $\sim 10^{-3}$ Gys^{-1} with an associated gamma component of ~6% of the total dose administered. The half life of the target under such conditions was ~50 hours.

Benchmark experiments

The neutron generator can be utilized for benchmark experiments relevant to fast reactor physics. Model blanket experiments have been conducted in several laboratories (see for example ref. 26) on neutron multiplication and tritium breeding. Other important fast reactor physics parameters could also be investigated.

Miscellaneous

Emission of recoils from (n,2n) reaction

The (n,2n) reactions have usually rather large cross sections ≥100 mb at E_n=14 MeV. The energy imparted to the recoils is sufficiently important (several hundred keV) so that some tens of radioactive recoils can find their way out of the target. Collecting them on a foil one can obtain data on the importance of the phenomenon, its dependence on various parameters like angle of emission, surface conditions elemental composition, crystalline form etc. Such measurements have not been too frequent in the past although some literature on the problem does exist[27,28,29].

The problem might be found of interest in solid state studies, for calculation of material losses in CTR's due to these effects.

Also in the study of (n,2n) reactions one might gather interesting data on the angular distribution of recoil nuclei. As far as I know, no literature data on the angular distribution of recoils exists so far, but there is no doubt that the described method could, with careful planning, yield interesting results.

Radionuclide production

14 MeV neutron generators could serve as a useful means for the production of some short lived radioisotopes[30,31]. Due to the fact that the cross sections for the 14 MeV reactions are relatively low and the fluxes available are several orders of magnitude lower than those available with nuclear reactors and cyclotrons, large quantities of material should be activated in order to obtain useful quantities of radionuclides. The irradiation of big samples is no problem, but it is important to develop procedures to separate the radionuclides from the

large bulk of target material rapidly and with high separation efficiency. Two factors assist in developing separation procedures. One is that the radioisotopes produced by (n,p) and (n,α) reactions differ chemically from the target material; the other is that the reaction energies involved are such taht the atoms undergoing nuclear reactions have a high probability of being torn from their parent molecules in the target. These factors have been shown to permit procedures based on elution of radionuclides from columns of irradiated material[31,32,33].

Experience has shown that several potentially interesting short lived medical radioisotopes like ^{13}N, ^{18}F, ^{64}Cu, ^{62}Cu, ^{63}Zn, ^{68}Ga and ^{78}Br can be produced for tracer chemistry experiments. The activities, however, are not carrier-free and their total quantities produced are too small to be able to label organic molecules for applications in humans.

Induction of mutations

Sporadically one finds in the literature reports about the effects of low doses 1-10 Gy on plants. It has been shown that the height of plants, the weight of fruits, the number of flowers etc, vary with neutron irradiation of seed, bulbs or growing plants.

A more systematic study of the phenomenon at biology and agricultural departments is necessary.

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SMALL ELECTROSTATIC ACCELERATOR AS A TOOL FOR RESEARCH IN MATERIAL SCIENCE

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Abstract

Utilization of a small electrostatic accelerator in a specific branch of material science is discussed. Ion beam techniques to modify the surface of materials to improve such properties as ware, friction, corrosion resistance is illustrated with specific examples. Formation of metastable alloys and extension of solid solubilities are discussed. Use of Rutherford Basic Scattering Technique for dept profiling is discussed in detail.

1. Introduction

The theme of this meeting is "Alternatives to Research Reactors and the primary purpose of this meeting - and I am quoting - is to discuss the alternatives to research reactors for those developing countries starting nuclear science and technology programmes, but find the research reactors beyond their means". In my opinion, establishment of a research and training programme based on the use of small accelerators is one of the most cost-effective ways to provide the first major step for the infrastructure and training needed for any nuclear science and technology programme, especially if it is not nuclear energy oriented. Apart from providing training in measurement technology and experimental techniques in physical sciences, such a programme could encompass applied research in areas as diverse as material science, biology and agriculture, dosimetry, medical science, neutron physics, neutron cross section measurements and environmental sciences. I would go further and say that even for those research organisations who already have a research reactor based programme, an accelerator facility would act as a complementary facility and extend the range and scope of the nuclear science and technology programme as a whole.

In this meeting we have talks which deal with use of accelerators in some of the areas I have already mentioned. Although any particular type of an accelerator is a versatile tool, it - by itself simply - does not necessarily cater to the whole range of these areas of research. Thus the choice of the type of accelerator depends very much on the interests and long term plans of the nuclear science programme of a particular country.

In my talk I would like to bring out the utilisation of a small electrostatic accelerator in a specific branch of material science - where it is a unique tool. Now before I go further let me explain the two qualifying terms I have used i.e. "small" and "electrostatic". By small I mean accelerating voltage upto a few MV maximum, and generally 2 MV or less, and the word "electrostatic" is to be interpreted as high energy resolution and easy energy variability. Generally an electrostatic accelerator satisfies these to criteria, and although some components of the programme that I am about to describe/illustrate could be carried out by another type of accelerator - say a cyclotron - an electrostatic accelerator i.e. a Van de Graaff accelerator - single ended or tandem type - is the most suitable accelerator for such a programme. There are at least three talks at this

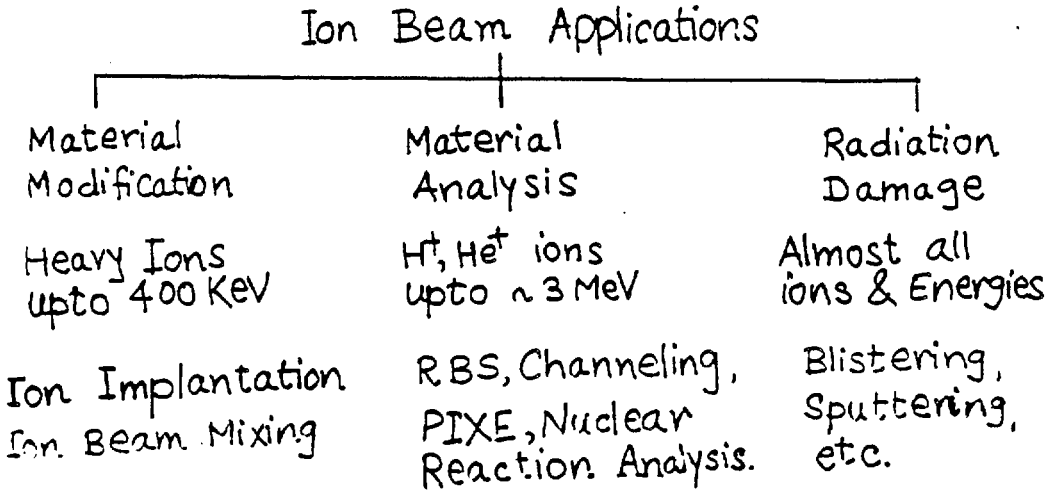
meeting which would deal with programmes which can be implemented through this type of accelerator which strengthens the case for such an accelerator. There is also likely to be some overlap and repetition between my talk and those other talks but hopefully just sufficient to stimulate your interest and little enough not to bore you.

Now before I get into the subject matter of my talk, let me explain that I am not a material scientist. I am primarily an experimental nuclear physicist, and secondarily an accelerator physicist in the sense that in all my professional life of 33 years I have dealt with various types of accelerators - have designed them, fabricated them, installed them, operated and maintained them and used them in my work in nuclear physics, and participated in or supervised programmes utilising them in other areas of physics. I am going to illustrate my talk with most of the examples from the current programme at the Van de Graaff laboratory at B.A.R.C. with which I was associated for the last 20 years till I came over to the IAEA last year. My own association with this work was mainly in the supervisory capacity as the head of the laboratory and not through direct participation except for very early stages. The programme is currently carried out by a very small group of young men led by Dr. Animesh Jain. Dr. D.K. Sood, who is no longer associated with this group was the earlier leader. I would like to acknowledge at this stage the considerable help I have received from Animesh Jain in preparing the part of the talk that follows. I hope that you do not ask me detailed questions related to metallurgy and solid state physics, on the other hand I would be glad to answer any question on the accelerator aspect of the programme.

To demonstrate that I am not narrow minded (!) and do not want to hark on the word "electrostatic" accelerator all the time, we will broaden the title and call it "ion beam applications in material science research".

We shall first start by describing the scope of such a programme outlined in terms of three areas of application of ion beams in material science as shown in Transparency 1. The range of energy of the beams and the techniques used for each area are also indicated.

I will deal with each of these areas, describe the technique and show some result.



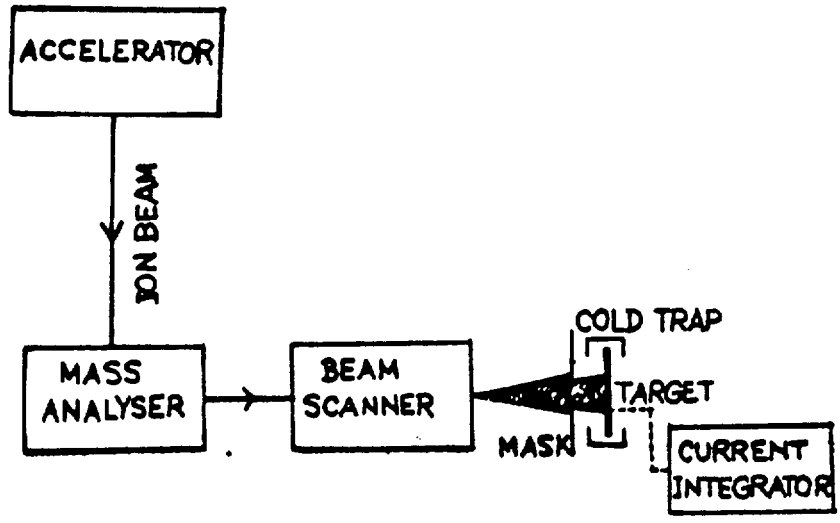
Tr.1

2. Material Moficiation

2(a) Ion Implantation

Let us start with the first one i.e. Material Modification through Ion Implantation. One uses beams of all types of ions for this purpose generally called "heavy ions" (in contrast with "light ions" which mean protons, deuterons, tritons, helions and alpha beams). The energy is upto 400 keV. The ions are implanted in the surface of the material - range of 400 keV heavy ions in material is very short (< 1 μm) and one is only dealing with surfaces here as against the bulk. The general term used for this technique is "Ion Implantation".

The schematic experimental arrangement for ion implantation is shown in Transparency 2.



Schematic of an Ion Implantation set-up

Tr.2

In the next Transparency (Tr. 3) I have indicated the special features and advantages of ion implantation over conventional alloying techniques in tailoring the surface properties of materials.

The ion implantation technique to modify the surface properties of materials is used both for metals and non metals. I will now describe some work done on metals at B.A.R.C. and some of the other laboratories.

ION IMPLANTATION

A technique to introduce controlled amounts of an impurity (or solute) into any target material.

Advantages

1. Cleanliness (in vaccuum $\lesssim 10^{-6}$ Torr)
2. Purity of alloying (Mass analysed beam)
3. Precise control & Reproducibility (elect. control)
4. Tailoring of surface Properties, independent of bulk properties
5. Economic use of Precious & Rare elements.
6. Tailoring of Dopant Profile (Multi-energy Impl)
7. No bulk Heating (Solid State alloying)
8. No dimensional change (can be used on finished components)
9. No problem of delamination or peeling, commonly encountered in conventional coatings (Does not produce a "coating")
10. A non-equilibrium process. Exotic "metastable" alloys may be formed which can not be produced by conventional techniques.

Examples: i) Extension in Solid Solubilities
ii) Amorphous alloys (Met. glasses)

Tr.3

Ion implantation in metals is carried out for either both or one of the two purposes:

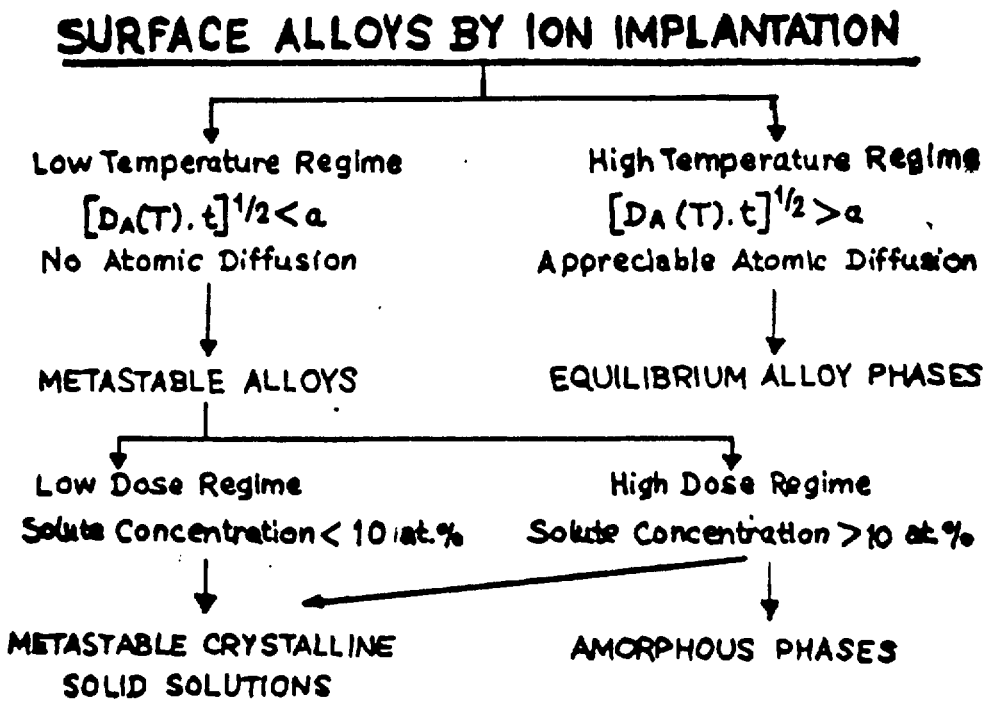
(1) Production of Metastable Surface Alloys (crystalline or amorphous)

(2) Tailoring of surface properties to reduce corrosion, wear, friction etc.

Metastable surface alloys are formed when atomic mobilities during implantation are low. This usually happens at or below room temperature. Even completely immiscible systems can be alloyed since the solute atoms cannot move and form precipitates during the process.

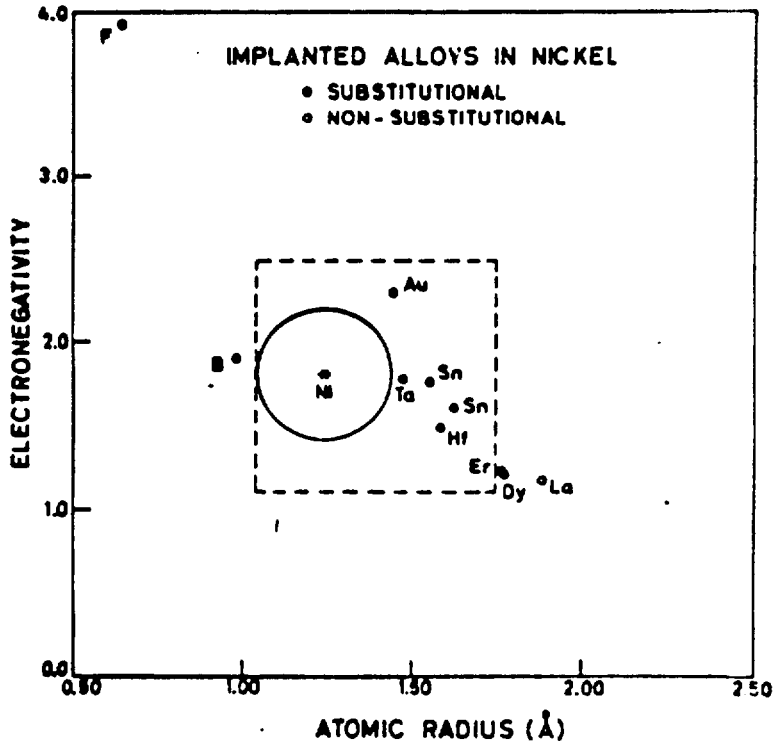
Metastable solid solutions (MSS) are formed at low doses (< 10 atom %). At higher doses transition to an AMOPHOUS PHASE is observed in some systems.

The surface alloys formed by ion implantation are classified as shown in the next transparency (Tr. 4).



Tr.4

In the next transparency (Tr. 5) I have what is called Darken–Gurry plots for implanted alloys in Nickel. The circle represents the region in which substitutional alloys are expected according to Hume–Rothery Rules. The data on dilute implanted alloys show violation of these rules in many cases as brought

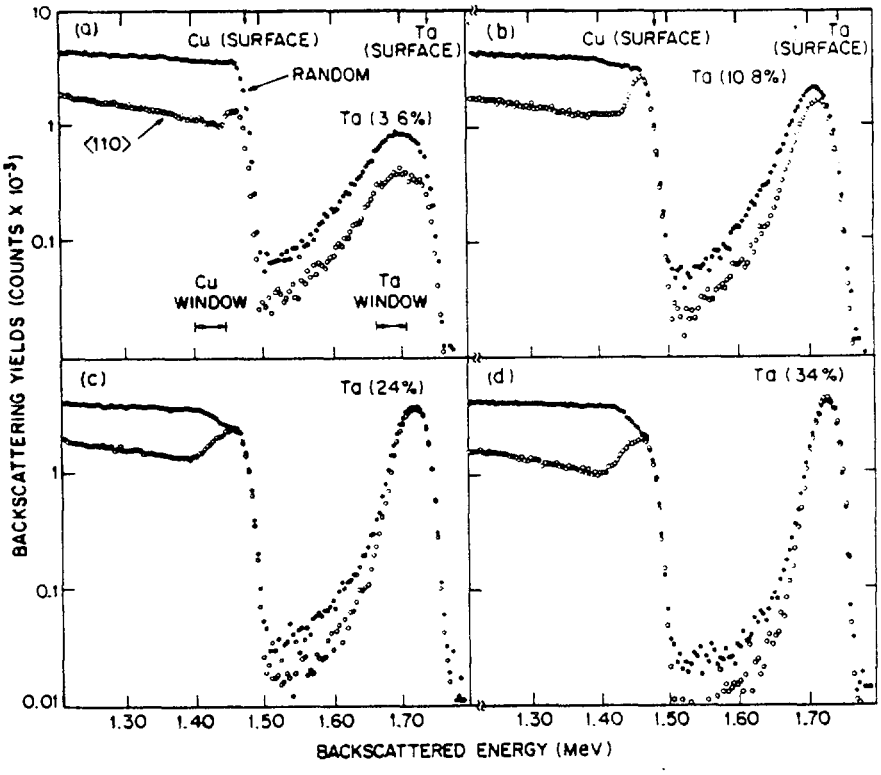


Darken-Gurry Plot for Implanted Alloys in Ni. Substitutional alloys expected only inside the circle. The dashed region depicts modified Hume-Rothery zone proposed by Sood (1978). Tr.5

out by work done at B.A.R.C.. Dinesh Sood, who did this work, has proposed modified Hume-Rothery zones according to which the implanted alloys are found to be substitutional in almost all the cases studied so far. The extension in zone of substitutionality is believed to be caused by the implantation process, thus permitting atoms with larger radii to occupy lattice sites which would have been otherwise not possible.

The last figure represented the final results of the experimental investigations. In the next transparency (Tr. 6) I would illustrate the techniques - R.B.S. - which is used to carry out these measurements. In that sense this is an illustration of the R.B.S. techniques and belongs to the general field of ion beam techniques used for material analysis - as against material modification - I shall discuss this more when we come to that section.

This transparency (Tr. 6) shows R.B.S. spectra for Ta implanted in Cu at various doses. These are "channeled" spectra as against "random" spectra. The fact that for low doses the tantalum peak is reduced in <110> direction compared to "random" shows channeling and indicates "substitutional" alloy. For high doese - lower two spectra - there is no reduction from "random" direction which indicates transition to the amorphous phase. The "tail" on the low energy side of the tantalum peak indicates depth distribution of tantalum into the sustrate of Cu.



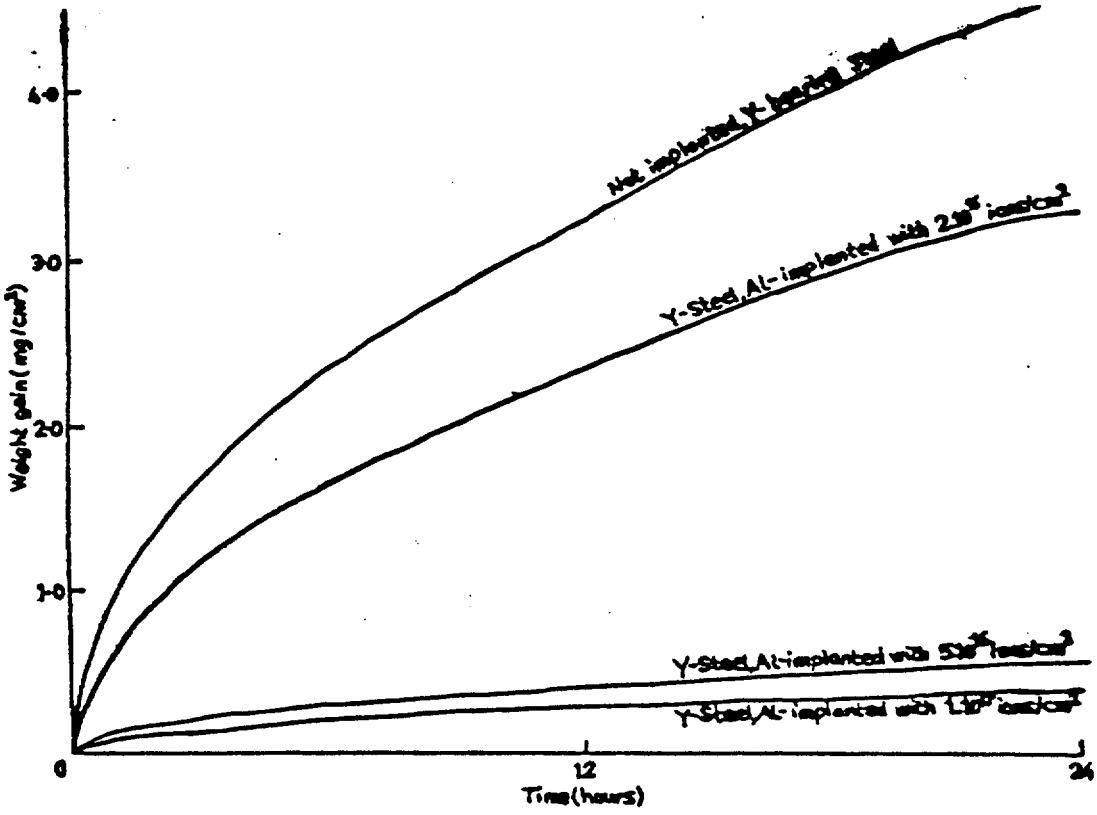
Random and Channeled <110> backscattering spectra for Cu single crystals implanted with Ta ions to peak concentrations ranging from 3.6 to 34.0 at.%. Transition to amorphous phase also confirmed by TEM.

A.G. Cullis et al
Philos. Mag. 37, 615 (1978)

Tr.6

Let us now see the use of implanted ions to modify material (surface) properties, as shown in transparency (Tr. 7).

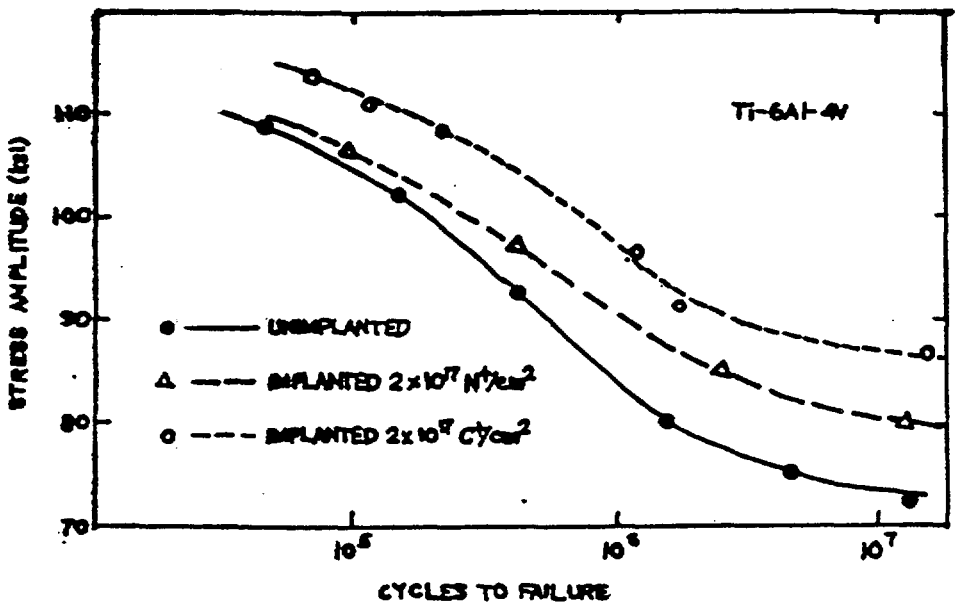
These are oxidation kinetics of Fe - Cr - Al - Y alloy at 1100° C for different doses of implanted Al from the work of Bernabi et al [Corros. Sci. 20 (1980) 19]. The enormous (factor of 140) improvement in corrosion charcateristic as a function of implanted dose is quite striking. The Al cannot be directly alloyed into the bulk because it would reduce duclibility of the material.



The oxidation kinetics of an Fe-Cr-Al-Y alloy at 1100°C in air for different doses of implanted Al (From Bernabai et al, Corros. Sci. 20 (1980) 19). The alloy contained only 1.45 % Al.

Tr.7

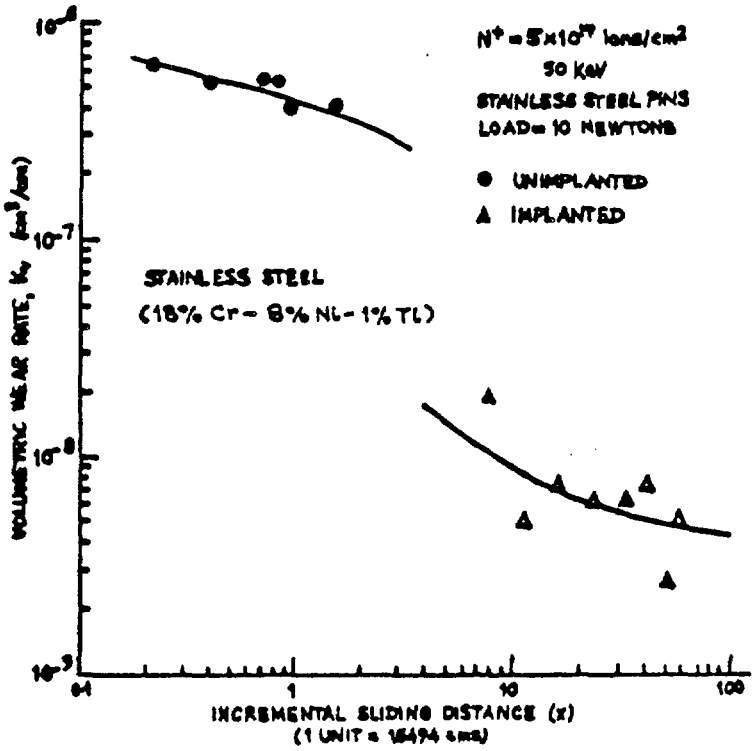
Similarly the improvement in fatigue endurance of titanium Al alloy implanted with carbon and nitrogen ions is illustrated in Tr. 8. This is from the work of Vardiman et al. reported in NRL report May 1979, USA. This alloy has application in making hip joint protheses for medical implants (in the body) due to its light weight and high strengths. It is seen that C^+ ions are more effective than N^+ ions. The improvement is due to formation of TiC particles.



The fatigue endurance of a Titanium alloy implanted with Nitrogen or Carbon ions (Vardiman et al., Report of NRL Progress, May 1979, p.4)

Tr.8

Similarly reduction in wear produced by N^+ implantation is illustrated in Tr. 9. The volumetric wear rate for stainless steel on stainless steel (loaded pin and disk method) improved by almost two orders of magnitude after implantation of 5×10^{17} nitrogen atoms/cm^2 at 50 keV.



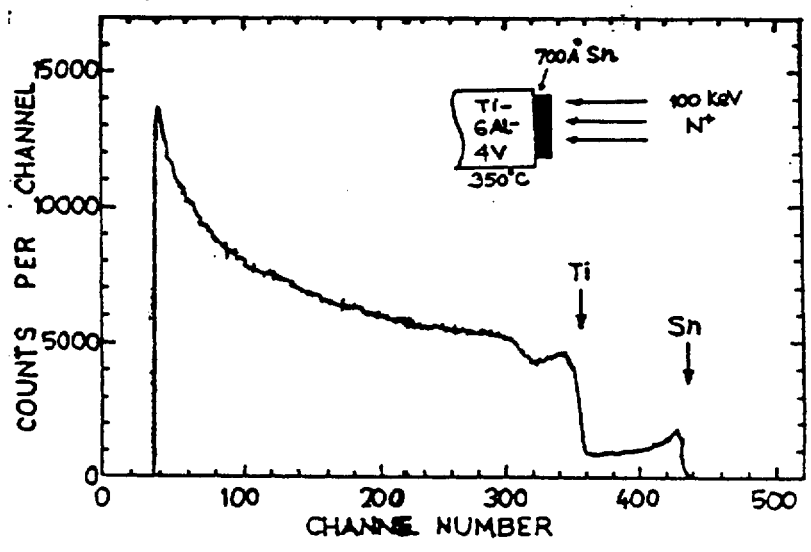
Volumetric wear rate of Stainless steel worn against itself in a pin-on-disc test.

[Dearnaley & Goode, Nucl. Instrum. & Meth., Vol. 189, p. 117 (1981)]

Tr.9

2 (b) Ion Beam Mixing

I would like to briefly describe a topic which was listed under "material modification" in my first transparency but have not dealt with it yet. The maximum concentration that can be achieved by implantation is limited to less than 20 atom % due to sputtering losses. Ion Beam mixing is a technique used to overcome this limitation. A thin film of the solute species is deposited (or implanted) on the substrate and then the surface is bombarded with energetic innert gas ions (N or Ar) or solute ions. If the film thickness is approximately equal to the range of bombarding ions, a collision cascade near the surface causes mixing of film and substrate material. This results in high concentration of surface alloys. A further refinement of this technique called "Bombardment Diffused Coatings" is when the ion beam mixing is carried out at higher temperature. Tr. 10 shows a RBS spectrum from a sample prepared be depositing a 700 A° thick Sn film on a Ti-6Al-4V substrate maintained at 350° C and bombarded with 100 keV N^+ ions. The spectrum indicates diffusion of Sn into the alloy due to radiation enhanced diffusion. This technique has the following advantages 1) high concentration, 2) large depts and 3) no need for solid charge ion source with high intensity. Only gaseous ions which can readily be obtained are needed. This is a great advantage for industrial applications.



Tr.10

Dearnaley Rad. Eff. 63 (19

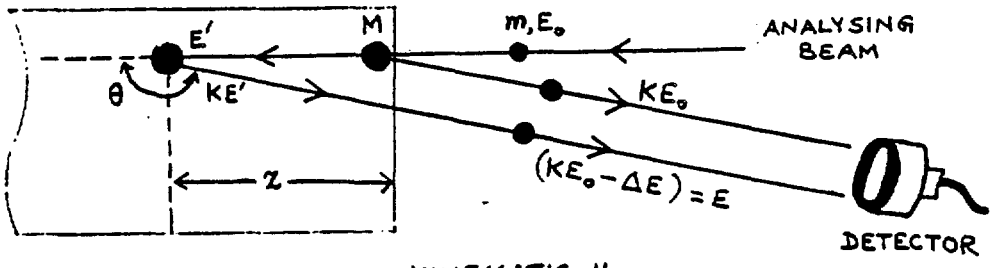
3 - Material Analysis

Let me now go over to the use of ion beams for material analysis. This by itslef is a very wide area and one could have an entire conference devoted to this. In this meeting we have talks on proton microprobe, PIXE etc. In my talk I want to restrict myself only to the technique used in material science. Depth profiling is a very powerful diagnostic tool in surface physics. There are two different classes for these techniques based on use of particle beams: destructive and non destructive.

The Auger Electron Spectroscopy and Secondary Ion Mass Spectroscopy (SIMS) are essentially sputtering techniques and therefore classify as destructive. Ion induced x rays, Nuclear Reactions and Backscattering Spectrometry depend essentially on energy loss principle and are non destructive techniques. I have already mentioned the Rutherford scattering techniques and show RBS spectra as examples of diagnosis after ion implantation. I would like to now explain the principle of the technique and show you some more examples of how the technique is used as a powerful diagnostic tool.

In this transparency (Tr. 11) the principle of Rutherford Backscattering (R.B.S.) technique is illustrated, schematically. The analysing beam (generally a 2 MeV alpha particle beam) of mass m and energy E is backscattered through an angle θ by coulomb interaction with the metallic atom M located at the surface of the sample with energy KEo where K, the kinamatic factor is a function of M, m & θ. An atom at a depth of Z units will scatter an alpha particle through the same angle θ with an energy KE' where E' is the energy of the alpha particle striking the atom at the depth Z having undergone a loss of energy equal to E_o-E'. The spectrum as seen by the detector (generally surface barrier detector) at angle θ is shown at the bottom. The figure is self explanatory. Typical overall energy resolution - beam energy spread, detector, electronics - has to be 10 keV or less for good quantitative results. The types of information deduced from such spectra are indicated as (a), (b) & (c). In addition comparison of "random" and "aligned" (channelled) yield provides information about lattice site location.

RUTHERFORD BACKSCATTERING



$K \equiv K(m, M, \theta)$: KINEMATIC FACTOR $\|$ $\Delta E \propto Z$ (depth)

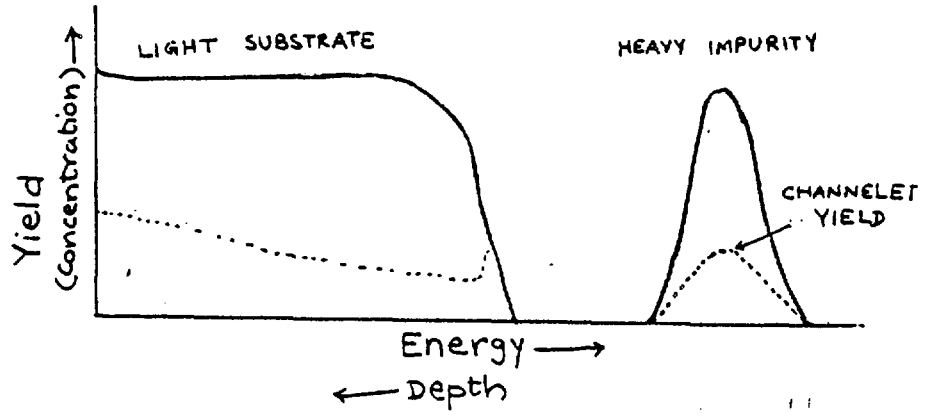
Yield at Energy E $\propto$ Concentration at depth z

a) K provides mass identification (m, θ fixed)

b) Energy loss provides depth information:

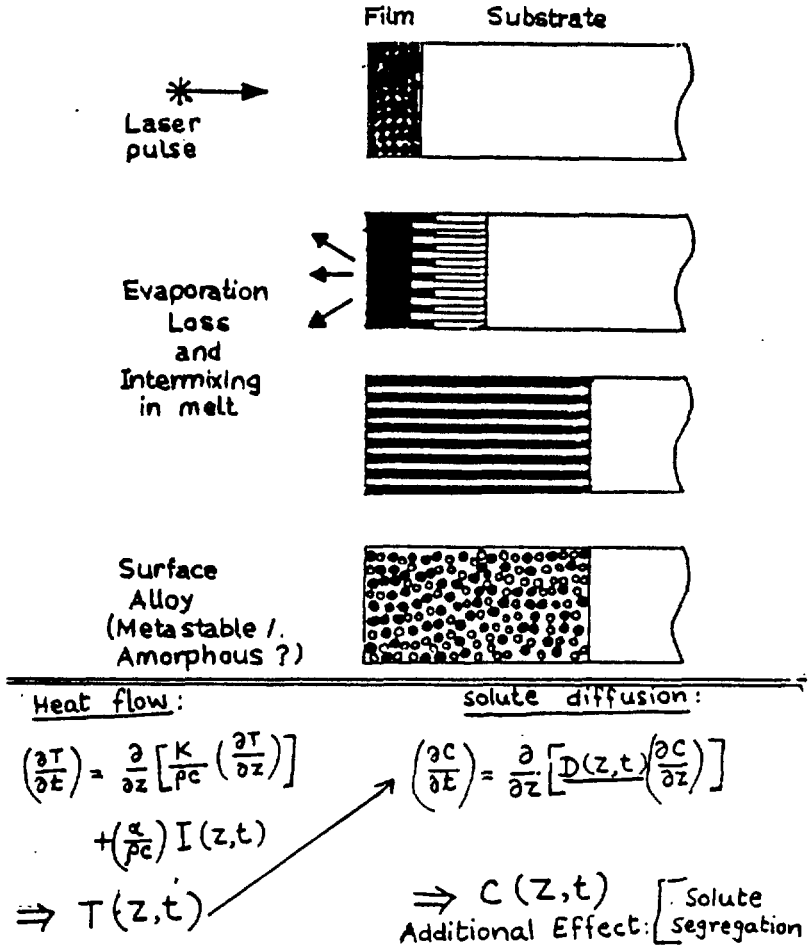
$$\Delta E \approx \left[K\left(\frac{dE}{dx}\right)_{E_o} + \frac{1}{|\cos\theta|}\left(\frac{dE}{dx}\right)_{KE_o}\right] Z$$

c) Yield directly related to concentration.



Tr.11

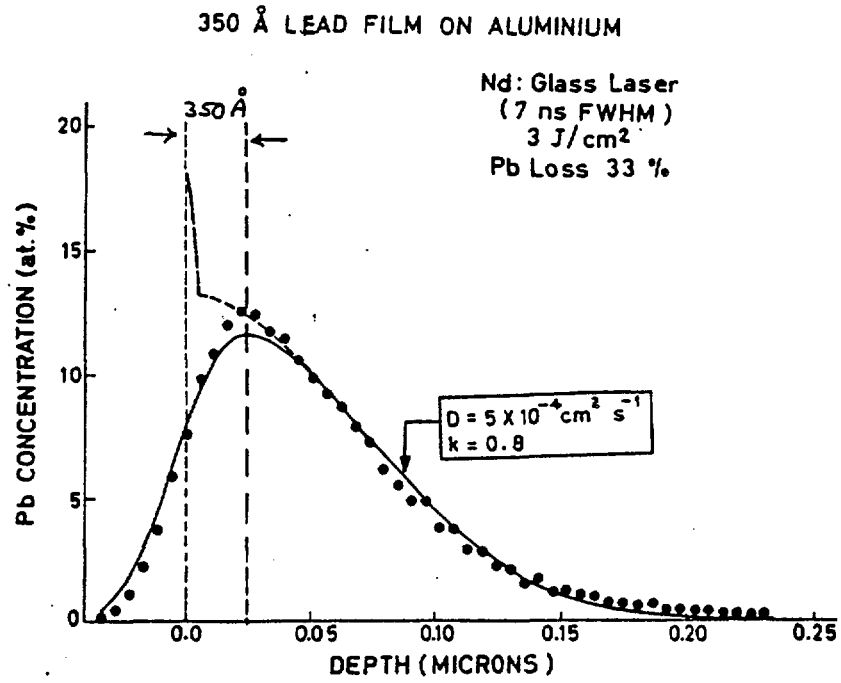
Now I would like to illustrate the use of RBS in the study of laser treated heavy metal films on light metal substrates. This brings me to the topic of surface alloys formation by pulsed laser treatment of deposited films. The films can be deposited on the surface by any of the standard techniques or implanted at a specific depth using ion implantation techniques. The diagnostics of the laser treated films can then be fully investigated using RBS and other surface analysis techniques. What is of relevance to this talk is the information one obtains through RBS techniques. But before I show that let me show in the next transpareny Tr. 12 schematic of the process of surface alloy formation by pulsed laser treatment of deposited films. Due to extremely high cooling rates $\sim 10^{9o}$K/s' (estimated from heating calculations) and very high resolidification velocities ~10 m/s, the final alloy is expected to be metastable. This method was proposed by the BARC group in 1979 and has been applied to several systems. Laser heating and solute diffusion calculations have been performed to understand the experimentally obtained solute depth profiles.



Tr.12

When such a film is subjected to full detailed RBS analysis the type of information one gets is illustrated in Tr. 13.

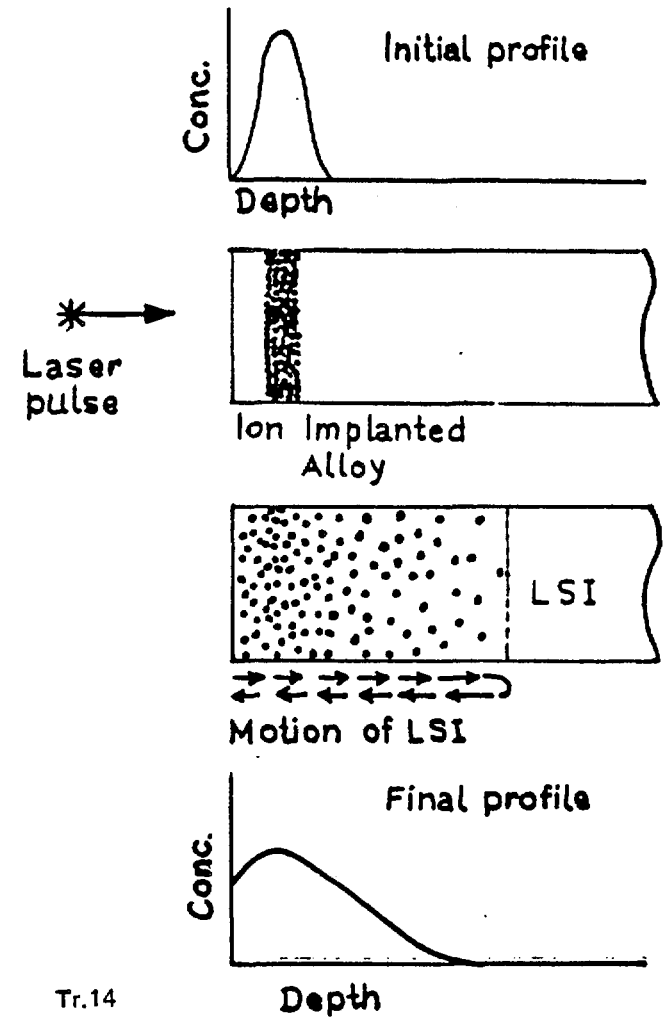
This is an example of application of RBS to study of laser treated Pb films on Al. The calculated curve is obtained by diffusion calculations. The initial profile is allowed to diffuse in a time-dependent melt depth. The melt-depth vs time information is obtained from heating calculations. Further, rejection of Pb into liquid at the time of solidification is also included via a segregation coefficient k, in the calculations. The diffusivity & k are varied to get best fits to experimental data. This work is again from BARC by Animesh Jain et al. first published in Aarhus conference on Ion-Beam analysis 1979.



Jain et al, Rad Eff. 63 (1982)

Tr.13

Laser treatment of an Implanted Alloy

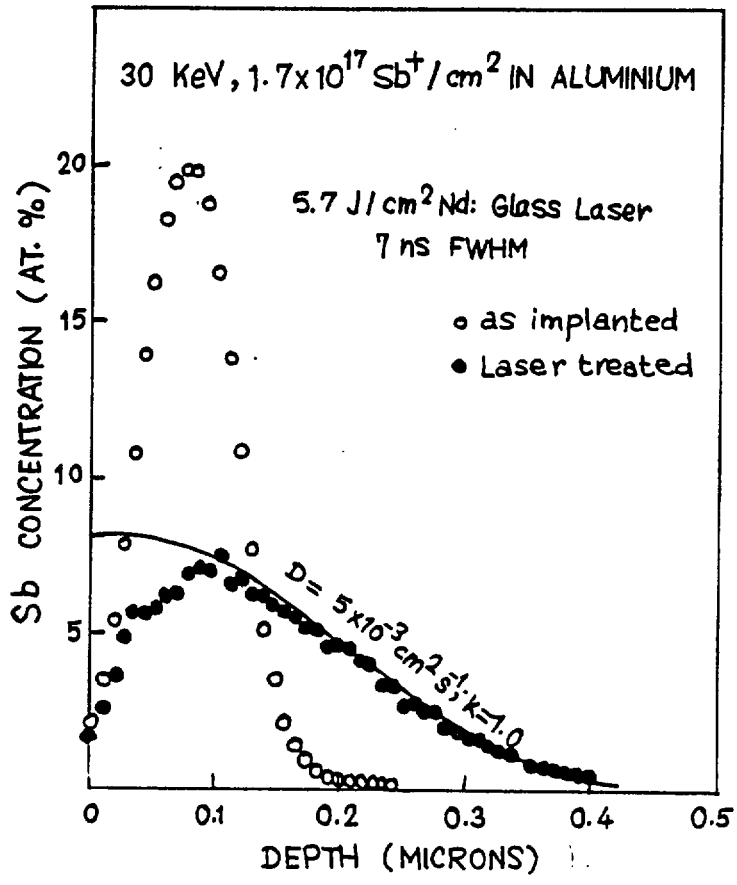


Tr.14

The earlier two figures are exmaples of laser treatment on films deposited by conventional techniques. The use of ion beams here is only for the diagnostics and it can be seen how invaluable it is.

We shall now look at laser treatment of implanted alloys. Here the ion beams are used to implant the foreign substance, and then the sample is pulsed laser treated. The next transparency (Tr. 14) is a schematic of laser treatment of implanted alloys. Advantages: i) Extension of alloying depth and ii) annealing of implantation damage, while still retaining metastable nature.

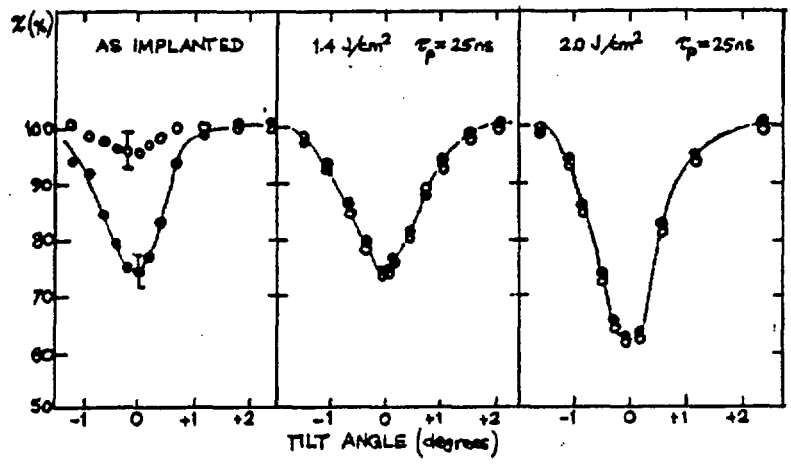
The full RBS diagnostics results on this sample are shown in the next Tr. 15 which is an example of depth profiles before & after laser treatment in Sb implanted Al. The calculated curve is obtained from similar calculation as in Fig. 19 for Pb/Al. The discrepancy between calculated and measured profiles near the surface is due to evaporation loss of Sb, which is not considered in the calculations (BARC work Animesh Jain et al.)



[Animesh K. Jain et al, Rad. Eff., 63, 183-185(1982)]

Tr.15

In the following transparency (Tr. 16), we have RBS spectra (channeling angular scares) for studies of Cr implanted in Al after laser treatment. The substitutionality in as implanted phase is absent as indicated by very shellow dip in the Cr yielded (open circles) in the left hand picture. The coinciding of open circles (Cr yield) with dots (Al-host yield) in the centre and right hand pictures indicate full substitutionality attained after laser treatment.



• Host yield (Al)
o Impurity yield (Cr) } <100>

Substitutionality Enhancement by Laser Treatment of an Implanted Alloy.

G. Battaglin et al,
J. Appl. Phys. 53 (1982) 3224.
(BARC - PADOVA work)

Tr.16

4. Radiation Damage

I had planned to talk a little about the third branch shown in my first transparency namely Radiation Damage. As we all know this branch of material science is very important in nuclear energy technology. I wanted to illustrate only one particular use of energetic ion beam - 2 MeV alpha beam - in the study of blistering and sputtering of surfaces under ion bombardment. The relevance of such research is directly with behaviour of the inner surface of the "First Wall" in a probable fusion reactor.

We carried out at BARC some time ago studies on blistering and exfoliation caused by bombardment of different materials under 2 MeV He^+ bombardment. The studies led to the proposal of a model by Deb & Sood for blistering exfoliation which considers the role of beam heating. Unfortunately I have run out of time and must stop here.

5. Conclusions

I have tried to bring out - perhaps in more details than necessary - the importance of a small accelerator as a tool for research in material science. I have not yet dealt with required specifications for the accelerator, and the auxiliary equipment necessary. A Van de Graaff - Tandem - type accelerator with a terminal voltage around 2 MV with a separate ion implanter with 400 KV voltage and multiple choice ion beam source would be an ideal combination. However, a lower voltage machine with more versatility could also equally well serve as a starting facility. The advantage of such a facility is that it is upgradable either upward vertically in terms of higher energies, or horizontally in terms of more variety ion beams and diversification in the areas of research by adding appropriate auxiliary equipment. I have not dealt with the nuclear reaction techniques used in material analysis nor techniques like PIXE, charged particle activation or thin layer activation by heavy ion beams, and associated studies in agricultural sciences, environmental sciences, biology archeology etc. Some of the other talks here have already dealt with a few of these applications. I do hope that I have made my case.

FAST HEAVY ION INDUCED DESORPTION OF BIOMOLECULES – PLASMA DESORPTION MASS SPECTROMETRY (PDMS)

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Abstract

Fast heavy ions like fission fragments from a ^{252}Cf-source induce desorption of molecular and fragment ions when they hit the surface of a bioorganic solid. This effect can be exploited in the ion-source of a mass spectrometer. As compared to other ionization methods available in mass spectroscopy this method is particularly powerful for large thermally labile biomolecules, e g proteins. A new area of research is rapidly developing. It involves studies of mechanisms for fast ion induced desorption and biomolecule ion formation and the development of experimental tools to determine the mass of these large molecular ions. Both ^{252}Cf fission fragments and beams from several heavy ion accelerators are used in this research. The work at Uppsala in this field will be briefly reviewed.

1. Introduction

The use of slow ions ($v<v_0$, v_0=the Bohr velocity) for surface studies is a well established field of research. Many phenomena in this field already have important technical applications, but are still the subject of basic research. The application of sputtering in SIMS analysis is a famous example. Another important use of slow ions is for modification of bulk properties of solids i e ion implantation.

Very few experiments where fast ions are used in SIMS measurements have been described in the literature. This is mainly due to the fact that until recently only slow ions were believed to cause significant erosion effects in solid surfaces. A few years ago it was found that also in the

electronic stopping regime dramatic erosion effects may occur and electronic sputtering has now been observed in insulators like alkali halides (1), in condensed gases (2), ice (3), metal halides (4) and in bioorganic solids (5,6). This means that SIMS studies can also be undertaken with fast ions. Already in 1974, Macfarlane and corworkers (7) used fission fragments from a ^{252}Cf- source to cause desorption and ionization of biomolecules like amino acids and peptides. It was found that this is a very efficient way to produce gas phase ions of labile molecules. By combining this ionization method with the time-of-flight (TOF) technique, the Texas group constructed a new type of mass spectrometer (8) and since then this group has studied a number of important and "difficult" biomolecules (9). The method was given the name Plasma Desorption Mass Spectrometry (PDMS) by Macfarlane and coworkers. The mass spectrometric results of Macfarlane and coworkers and the observations independently made at Uppsala (5) and Erlangen (6), that a fast ion erodes a bioorganic solid surface have opened up an entirely new field of research. It ranges from mass spectrometric studies of molecules like protected oligonucleotides (9) to investigations of the features of the ionization method itself, like the importance of metastable ion-decay processes (10). Furthermore groups at several accelerator laboratories are now studying the mechanism for fast heavy ion induced desorption of biomolecules (5,6,11,12). The successful use of fast ions for producing gas-phase ions of large and thermally labile molecules can be illustrated by the recent observation by our group of quasi-molecular ions from a 119 amino acid residue protein of mass 13309 (13). Further references to studies in this field can be found in the proceedings of workshops on this subject held at Uppsala 1981 and Münster 1982 (14).

The use of fast ions for SIMS analyses of bioorganic solids is by now well established as was illustrated above. It may be added that it has recently been shown that fast ions can also be used for material modification purposes (15).

After Macfarlane et al started to use fission fragments to induce desorption of biomolecules from a sample surface, Benninghoven and coworkers (16) applied the SIMS method to the study of bioorganic solids. With the use of a 2 keV Ar^+ beam, they were able to obtain spectra of amino acids, spectra which were very similar to those published by Macfarlane et al (7).

Another development of the SIMS-method is the use of a liquid matrix, e g glycerol, in which the biomolecules are dissolved. This technique has been given the name FAB (Fast Atom Bombardment) by its inventors Barber et al (17). The FAB-method has recently been used to produce quasi-molecular ions of bovine proinsulin, a protein of mass 8687 (18). The fast ions seem to give higher yields, i e the number of moelcular ions per primary ion, for desorption of large molecules and, it is a very exciting field of research to try to find out the relative merits of fast and slow ions as desorption tools. Some studies have already been made (19,20) but this is an area where more research is needed.

2. Time-of-flight mass spectrometry of proteins

A time-of-flight mass spectrometer of the same type as that originally proposed by Macfarlane et al (8) based on the use of fission fragments from a ^{252}Cf-source has been used to obtain mass spectra of positive ions from proteins. The principle of the spectrometer is shown in fig 1. The details of the experimental technique used in this study will be described elsewhere (21). The main features are the following. The sample molecules (≈100 µg) were dissolved in triflouracetic acid and electroprayed (22) onto a thin aluminium foil. The fission fragments pass through the Al-foil and cause desorption and ionization from the layer of biomolecules. The associated fission fragment is detected in a start detector and initiates a time-to-digital converter (TDC). Keeping the sample at +17 kV the desorbed secondary ions are first accelerated and then allowed to drift in a

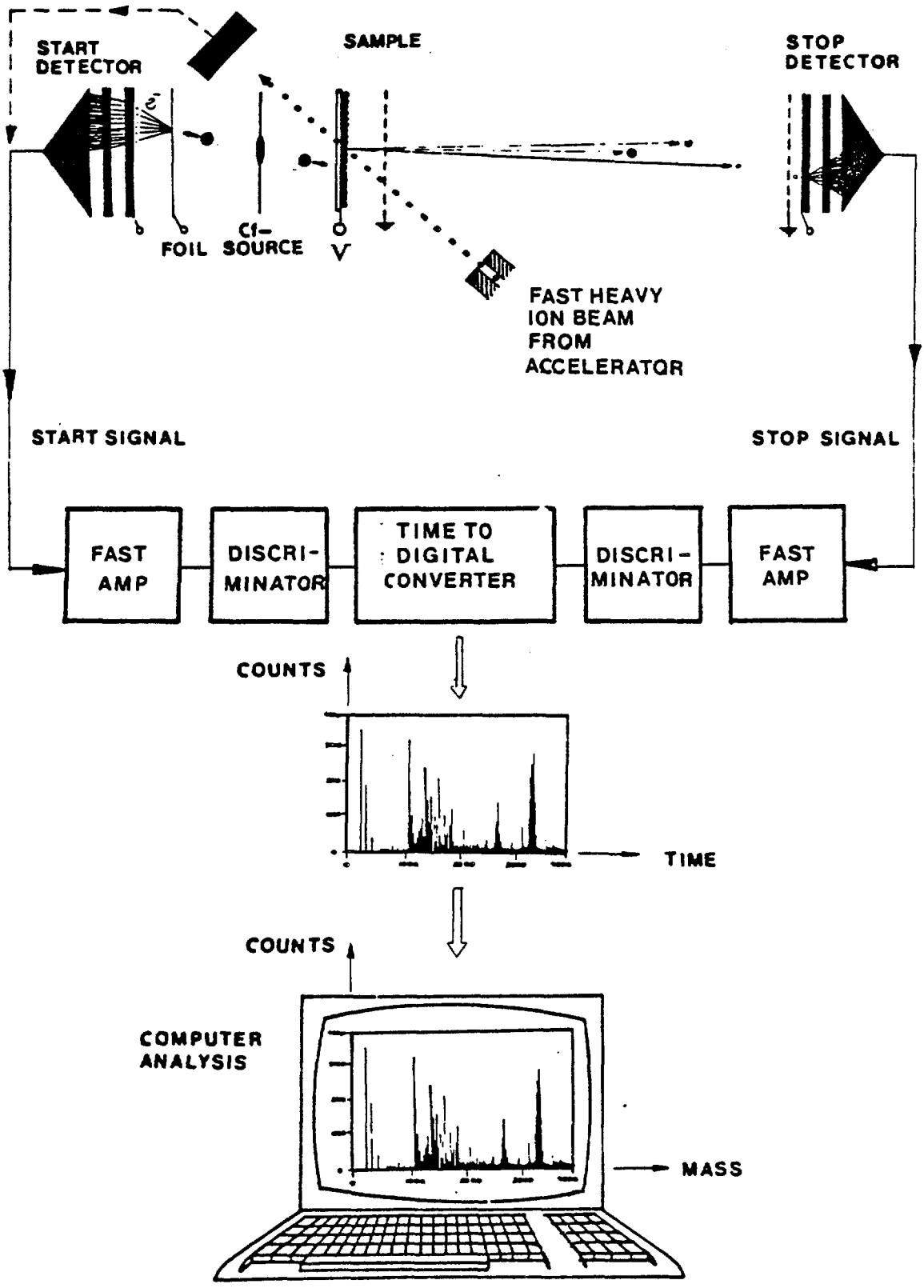

Fig. 1. The principle of the ^{252}Cf time-of-flight mass spectrometer used.

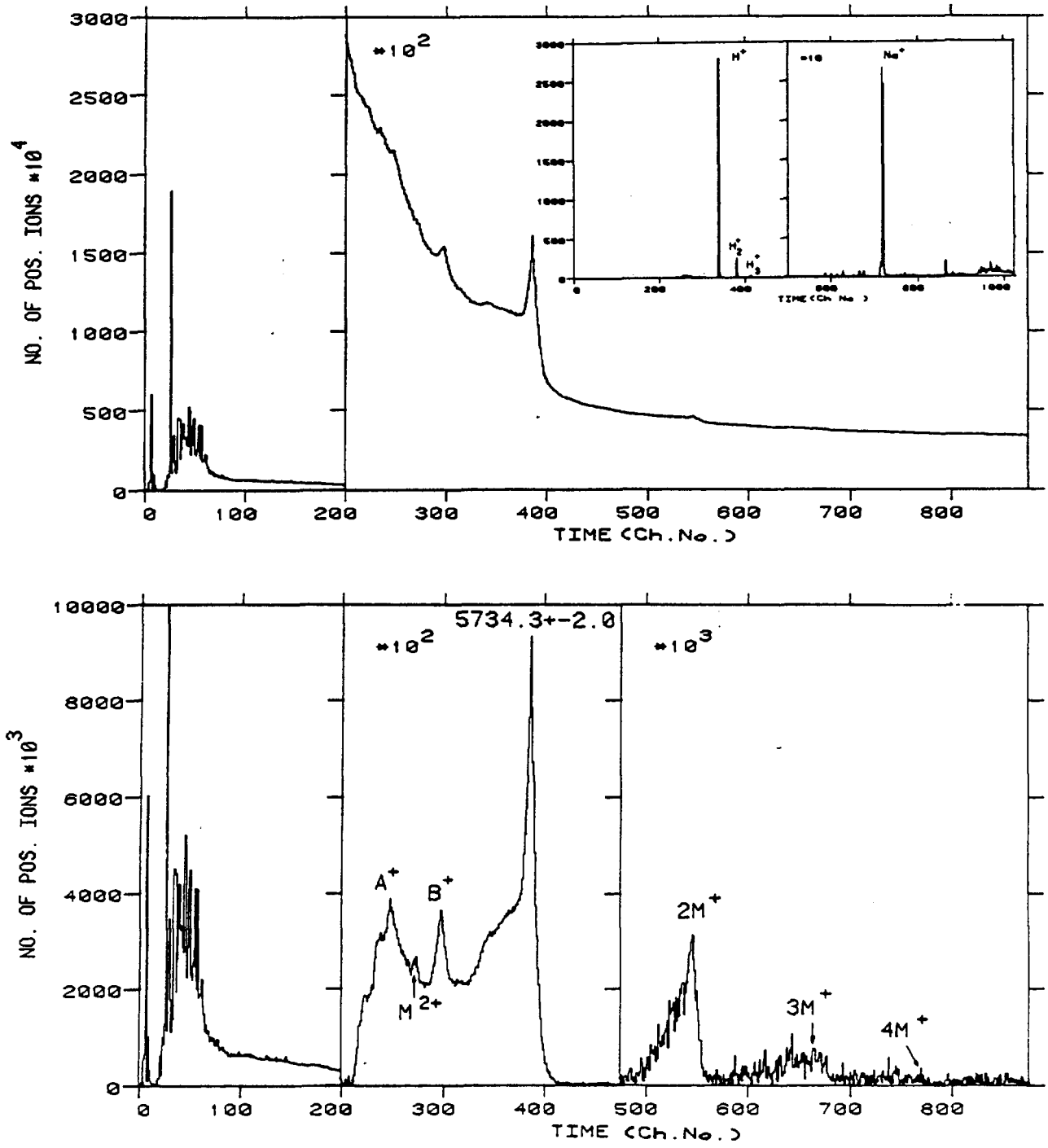

Fig. 2. The time-of-flight spectrum of positive ions from bovine insulin (MW 5733). (a) The total spectrum. (b) Background subtracted spectrum.

field free region before they enter the stop detector. each fission fragment induces desorption of several, often more than 30 secondary ions and it is important that the TDC can accept several stop signals per start signal. Time-of-flight spectra are collected with the use of a mini-computer. In fig 2a the raw spectrum of bovine insulin is

shown and fig 2b shows the same spectrum but with a smooth background subtracted. This background is most likely due to ions which decay in the acceleration region. In fig 3 the structure of this molecules is shown. It consists of two strings (A-chain and B-chain) of amino acid residues (21 and 30 respectively). The two chains are coupled together with disulphide bridges. The structure associated with the main peaks is indicated in fig 2b. The spectrum displays most of the features of TOF-mass spectra of proteins i e the quasi-molecular ion $(M+H)^+$ *, polymer ions ($2M^+$ and $3M^+$) and doubly charged quasi-molecular ions M^{2+}. This latter effect is even more clearly illustrated in fig 4a. This is the spectrum for porcine phospholipase A_2 which is a 124 amino acid residue protein of mass 13980 amu with seven disulphide bridges. In this spectrum there is a peak due to not only doubly but also triply charged quasi-molecular ions. In fig 4b the corresponding negative ion spectrum is shown. Also in this case multiply charged ions are observed ($M2^-$).

* M means here the isotopically averaged mass.

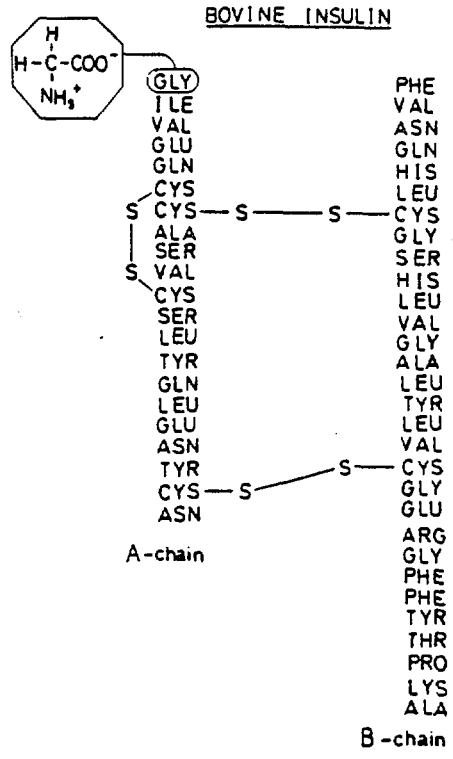

Fig. 3. The structure of bovine insulin.

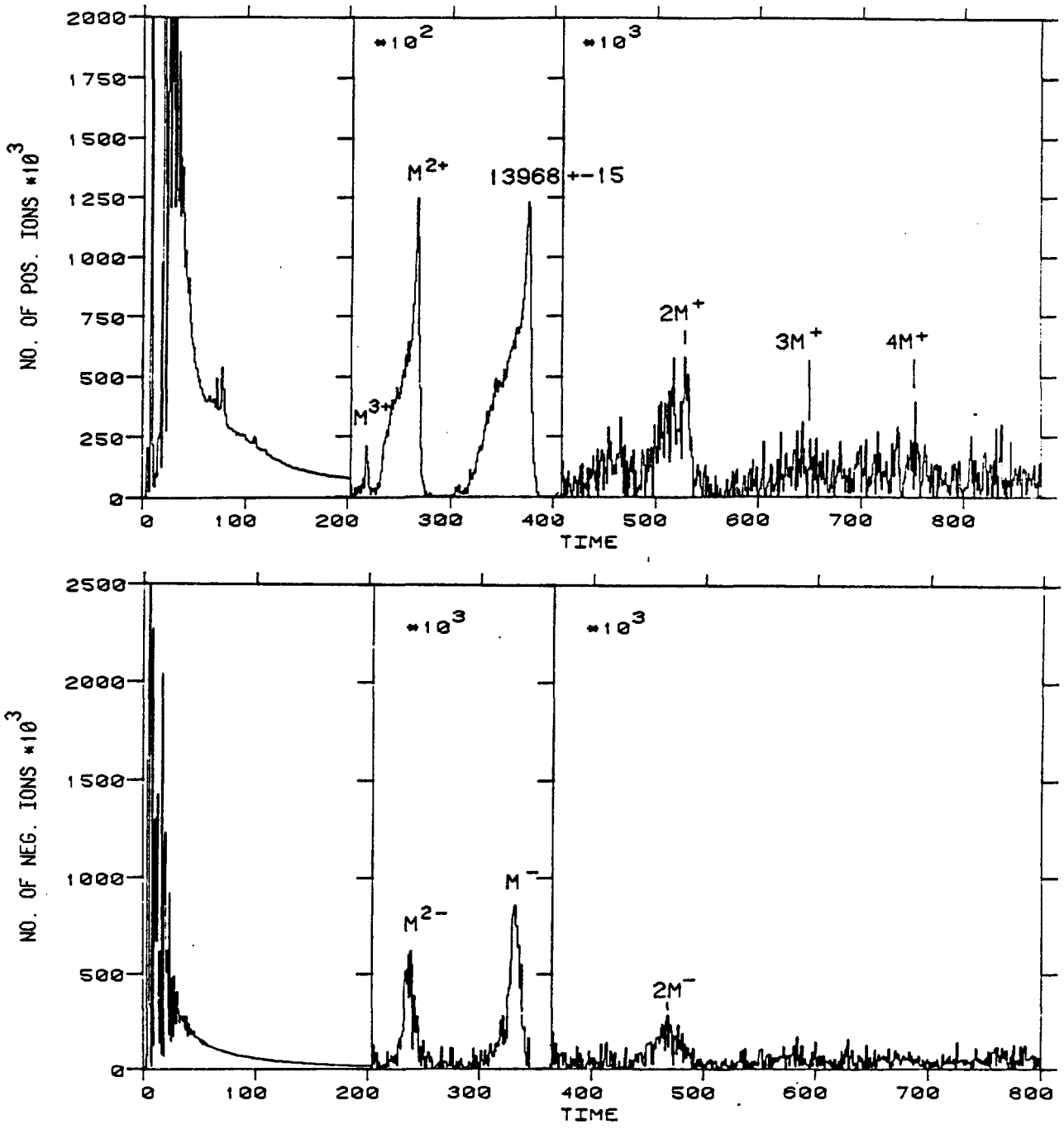

Fig. 4. The time of flight spectra of porcine phospholipase A_2 (MW 13980). (a) Positive ions. (b) Negative ions.

3. Final remarks

The field of fast heavy ion induced desorption of molecules is expanding very rapidly. It is a true cross disciplinary field involving scientists from many fields like physics, chemistry, medicine and the biosciences. At present there are three main directions of research. The application of fast ion induced desorption i e PDMS in the structure determination of proteins and other large molecules is a

completely virgin and promising area of research. The mechanism for conversion of electronic energy to molecular motion is a basic and important problem in the field of fast ion induced erosion. Research in this field can be performed with a system including a ^{252}Cf-source, a vacuum system and a small computer. It is therefore considered well suited for small department at a physics institute even a developing country. A system of this kind is at present beeing constructed at the Physics Department, University of Colombo, Sri Lanka in collaboration with the Uppsala group.

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SUMMARY OF DISCUSSIONS

1. Research reactors are versatile tools and find multipurpose utilization. A small reactor has cost advantages, both capital and operating, and can play a significant role in the introduction of a nuclear power programme.

2. Radioisotopic sources are relatively inexpensive and are within the means of most laboratories. They are small and conveniently calibrated. They may be used for calibration purposes and in spite of their low neutron yield, can be used in neutron physics experiments and some differential cross section measurements. The neutron spectrum from a ^{252}Cf source closely match those experienced from unmoderated fission neutrons.

3. Cyclotrons are extremely versatile machines and find multipurpose utilization. However, they involve sophisticated technology.

4. A small accelerator is a good machine for a country with limited financial and manpower resources for the introduction of nuclear science and technology. This machine could be considered a logical first step before the introduction of a research reactor or cyclotron.

The following sections discuss the principal characteristics of small research reactors and alternative machines in the approximate or lower cost range. While details are not presented regarding utilization programmes, it was felt that initial studies can be undertaken regarding choices of machines based on the discussions. To produce any greater detail was beyond the time limitation and manpower resources available at the meeting.

Research Reactors

Research reactors continue to be excellent facilities to support radiation research because of their relatively high available neutron flux, large irradiation volume and reliability. The available neutron flux levels at the research reactor dictate the possible areas of research. Fundamental research in material science generally requires higher fluxes. Isotope production for several applications and neutron activation analysis on a routine basis can be done at moderate fluxes in the order of 10^{13} n cm^{-2} s^{-1} or about 1 MW power level. In general the extent of the use of a research reactor depends on the scientific and technical infrastructure. Hence co-location with academic institutions/ education/research/industrial/medical centres is recommended.

A very significant contribution made by research reactors and their utilization is in the training of manpower tuned to the requirements for introduction of nuclear power in any country. It helps to prepare a sufficient number of scientists/engineers who are required even at the planning stage. After the construction of nuclear power plants, the research reactor continues to train operators and other operations and safety-related staff. The value of a research reactor for this purpose is increasingly realized.

The minimum cost of a small research reactor facility is of the order of 2-4 million dollars depending on the power level and variety of uses for which the reactor is designed. Major changes in the layout and savings in building and shielding costs could be attained if the research reactor is designed solely for the purpose of isotope production, training in reactor physics or activation analysis.

Small Accelerators

Accelerator based neutron sources using the D-T reaction have contributed significantly in initiating neutron based research, and thus familiarity with nuclear techniques, in a number of centres in both the developed and developing countries. An accelerating potential of about 400 kV and a few milliamps of beam current are minimum requirements. Such a facility could serve in initiating research in several areas on a modest scale similar to that in a research reactor centre. A special advantage could be that problems requiring fast neutrons could also be investigated. The cost of establishing such a facility is moderate (under 500,000 $) and the manpower needs are correspondingly small. The experience gained in nuclear technology is mostly limited to basic nuclear techniques and does not cover the areas of reactor kinetics, control and engineering systems. Nevertheless this could be an important starting point for a country with limited resources and capacity in science programmes at the post graduate level.

In recent years low energy ion beams have found extensive use in ion implantation, ion beam diagnostics and in analytical work. Electrostatic accelerators (up to 5 MeV) have been useful in carrying out innovative research in the above areas. Such programmes may lead to training in areas complementary to fission reactor technology.

Cyclotrons

Cyclotrons are extremely versatile machines and find interdisciplinary utilization. A medium sized cyclotron (E = 40 to 50 MeV) is useful not only for fundamental studies in nuclear physics but also in the fields of material science, chemistry, biology and medicine. Investigations of nuclear reaction theories and properties of nuclei away from stability, simulation of radiation damage in metals and alloys etc. and studies of radiation effects on chemical and biochemical molecules are some examples. Experimental measurements in the area of neutron physics can be extended to higher energies. Both monoenergetic neutrons in the energy range up to 20 MeV and intense fast neutron spectra can be obtained. Regarding more practical applications, cyclotrons are useful for therapy (with both charged particles and neutrons) and, above all, for production of short-lived medical radioisotopes. In short, utilization of such machines involves training and expertise in a vast number of areas similar to those covered by a research reactor. The demanded level of technology, however is higher and the costs involved are also higher (>3 million dollars). If the primary aim of a country is to develop nuclear science and technology associated with the use of radiation and isotopes, a medium sized cyclotron is an excellent alternative to a research reactor.

Isotope Production or Isotope Dependent Research

Apart from the already well established areas of research based on reactor produced isotopes, a great variety of short lived isotopes have become important for medical research and in-vivo diagnosis. The science of nuclear medicine is making rapid strides with the availability of short lived isotopes and with the use of modern techniques of detection and analysis. Thus gamma scanning, single photon emission computed tomography (SPECT) and positron emission tomography (PET) have enabled the study of both morphology and dynamical functions of various human organs. Except for ^{99m}Tc most of these isotopes are produced using accelerators. Proton, deuteron, ^{3}He and ^{4}He particle beams up to 100 MeV from accelerators (cyclotrons and Tandem accelerators) coupled with good nuclear chemistry facilities and imaging devices have been used. This has also led to the development of cyclotrons fully dedicated to medical use in the cost range of 2 million dollars. Introduction of methods in nuclear medicine in a country however demands a greater awareness and acceptance of a new science in that country. However, this is definitely a high technology area and requires an advanced hospital in the vicinity of the accelerator.

For more general biological applications, through in-vitro methods, the use of reactor produced tritium and ^{14}C is very common. Similarly the reactor produced ^{125}I is used extensively in radioimmunoassay. It seems therefore that in the areas of biological/medical applications, on a wide scale, the research reactor and cyclotron are complementary.

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