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Medical Cyclotrons *

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ABSTRACT

Cyclotrons as tools for therapy and for the production of radionuclides for use in nuclear medicine have been extensively reviewed in the literature. The current world status with respect to cyclotrons used primarily for research, development and application in nuclear medicine is reviewed here in the context of geographical distribution and type of use, presently available commercial types, machine characteristics and trends. Aspects of design requirements from a user perspective such as machine, beam and target characteristics are covered. Some special problems concerning many factors which can lead to effective production of the desired radionuclide or product are considered in light of machine characteristics. Consideration is also given to future directions for accelerators in nuclear medicine.

Introduction

The use of cyclotrons for medical purposes has been extensively discussed in the literature. The reader is directed to the extensive BNL

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bibliography on accelerator produced nuclides for use in biology and medicine for a comprehensive list of the literature prior to 1974 [1].

A review of the many national and international meetings on cyclotrons [2] would be beyond the scope of this paper, however, reference to some of the many papers available will be made. The perceptive review of Ter Pogossian and Wagner [3] gave strong impetus to the renaissance of the cyclotron in biology and medicine. While a number of research efforts in the biological and medical field using cyclotron produced isotopes were begun in the thirties in the U.S.A., a very sparse literature in this field exists for the forties and fifties. This is perhaps not surprising since the first cyclotron wholly dedicated to research and application in medicine was installed in 1955 at Hammersmith Hospital, London, the next wholly dedicated machine being installed at Washington University, Mallinckrodt Institute of Radiology in 1965. However, from the early sixties to the present day the number of papers and the range of applications in biology and medicine has been increasing rapidly. At the present time there are over two thousand papers in the literature concerning cyclotron use in nuclear medicine exclusive of applications in therapy and there are at least 61 cyclotron installations involved in full or part time programs.

A number of papers have appeared in the literature which review cyclotrons in medicine. A small number of these are listed [3-10] to give an overview of the various aspects of the problem. The exemplary effort of Chaudhri et. al. [6] in 1972 was the first attempt to relate cyclotron capability and cost in a critical manner. It is my own feeling that a careful analysis of machine parameters, what would be most desireable in light of current knowledge and what we can forsee in the future still remains to be carried out. The complexity of this problem is underlined

by the fact that there are perhaps four major areas to consider in terms of appropriate cyclotrons.

1. An integrated "small" cyclotron-imaging complex where the cyclotron has restricted capabilities in terms of numbers of nuclides which can be produced. Emphasis would be on automated or semiautomated production of nuclides and labelled compounds for daily use in conjunction with an on-line data collecting complex for routine use in diagnosis and patient management.

2. A "medium" energy cyclotron for daily production of a wider spectrum of nuclides for use in a large hospital or University hospital complex. Such a machine could also be used for basic research in nuclear medicine and its related fields.

3. A cyclotron in the higher energy range with little restriction on production capability solely for research and development in the nuclear medicine field, suitable for a University environment or large research institute environment where interdisciplinary and multidisciplinary work can be effectively carried out.

4. Cyclotrons [or other accelerators] solely used for large scale nuclide-production, primarily commercial in nature.

There is of course considerable overlap between the size of the machine and the uses defined by 2 and 3. Indeed the term "medium" covers a rather broad spectrum. It must also be explicitly stated that there are accelerators other than cyclotrons that can be used for research and nuclide production. Further comment on this problem will be given later. In a sense the proliferation of cyclotrons today is an example of technology outstripping basic science. Aside from the question of what would be most convenient for nuclide production and labelled compound preparation, cyclotron produced nuclides have not as yet impacted on health care to

the same extent as have the reactor produced nuclides notably technetium-99m, iodine-131, and a few others. Of the some 12 million doses of radionuclides used last year in the U.S.A. for medical purposes, only a small proportion were cyclotron produced, although some of these are indeed most useful in the delivery of patient care. Nevertheless, this symposium may indeed be related to what may be a turning point for the application of accelerator produced nuclides in the medical context. With the increasing sophistication of scanning devices with the increasing work on new detectors and methods, removing or at least reducing the restrictions of the past with regard to photon energy, and perhaps most importantly with the development of emission tomographs, the cyclotron produced nuclide may be coming into its own. The increasing availability of positron emitting nuclides and compounds labelled with these nuclides and their direct involvement in functional and metabolic processes opens up a whole new range of application in the "non-invasive" dynamic measurement of physiological and individual biochemical processes *in vivo*.

Cyclotrons - Types and World Distribution

There are basically two origins for the machines used in nuclear medicine today. The majority of cyclotrons designated as "medical" have been constructed by commercial manufacturers. The second class of machines are those whose original purpose was research in physics or chemistry or where these disciplines are the major thrust for use and a minor thrust may be use in biology or medicine. Some of the machines in this second class have been converted to full time nuclear medicine use. Many of these machines are of specialized design and were constructed at the particular university or institute.

No consideration was given to cyclotrons which are used wholly or in major part for particle therapy. Some machines listed do have therapy

programs as part of their regular schedule of use, however the list focuses on machines for chemical, biological, and clinical research in the nuclear medicine context and routine use in clinical application. While particle therapy is not appropriate to the intent of this paper it should be noted that an appropriately designed cyclotron can quite easily serve both purposes, nuclide production and particle therapy.

At the present time there are four active commercial companies engaged in cyclotron manufacture. These are the Cyclotron Corporation, Berkeley, California, USA, CGR MeV, Courbeville, France, Scandatronix, Uppsala, Sweden, and Japan Steel Works, Tokyo, Japan. There are numerous other organizations which have in the past designed and built cyclotrons for example, Philips (The Netherlands) and which should be considered to be in the field, however, to the authors current knowledge only the above four companies have as part of their active concern and sales the construction and delivery of "medical" cyclotrons. The types of machines available (Only machines which have actually been or are being constructed are listed) and their stated guaranteed design criteria, as given in the company brochures, are listed in tables I and II. It should be noted that guarantees on delivered current are frequently exceeded and in some instances the delivered energies are higher than guaranteed. For comparison purposes a listing of "large" cyclotrons (> 100 MeV particle energy) and accelerators used for medical nuclide research and production in a symbiotic relationship to the main purpose of these machines is given in table III

The geographical distribution of cyclotrons used full or part time for nuclear medicine is given in table IV and a listing of these machines is given in table V. The listing in table V does not however give an adequate picture of the situation. Machines which are listed under dedicated are indeed used full time in nuclear medicine research, however

there is considerable variation in application.

An institution may have any combination of extensive medical applications programs, clinical research programs, nuclide research programs radiopharmaceutical research and development programs, biological and medical research programs, instrumentation research and development programs, therapy programs, all centered about the use of their cyclotron. In reality very few if any institutions can be equally strong in all of these activities, yet they all constitute aspects of nuclear medicine research and practice. A similar but more restricted situation exists for the institutions listed under E.P. or established programs. A small number of these Established Programs are almost as extensive as those listed under D, but beyond this small number there is wide variation in commitment. The major portion of those under E.P. have nuclide research and production programs and a smaller number of this group have ongoing radiopharmaceutical research and development programs. Only a small number under this latter group have a real ongoing commitment to nuclear medicine. At the extreme end of this spectrum are those groups which produce nuclides only on a regular but limited basis with no commitment to nuclear medicine per se except to provide medically useful nuclides.

Cyclotron Characteristics and Size

In turning from the world situation as it exists at this point in time, it is appropriate to focus on what one looks for and expects in a cyclotron. Part of this question has already been addressed in the introduction and an extensive review of this subject is not the purpose of this paper. Nevertheless, one cannot divorce these considerations from a discussion of medical cyclotrons.

The use of a cyclotron in nuclear medicine research and clinical application is necessarily an inter-and multidisciplinary effort. The

primary use and development will involve physicists who design and operate these machines and chemists who must use the beams, do the appropriate research and prepare the nuclides and radiolabelled compounds. The biochemists biologists and physicians then use these materials in research and application. It is however far from being this straightforward. Overlap in interest and direction is very strong in this chain.

The physicist and chemist must consider what is most desireable in a particular demand context and what is feasible. The medical researcher must consider what is work limiting, quantity and/or type of cyclotron product.

Reviews of medical cyclotrons have been listed (vide supra). The chapter by Hoop, Laughlin and Tilbury [7] pages 407-419 can serve as an excellent elementary introduction to cyclotrons and nuclear reactions.

A cyclotron can be evaluated in terms of the machine and beam characteristics. Reliability and simplicity of operation are paramount. Reliability is self-evident if it is to be used for routine medical purposes. Should a multiparticle machine prove ultimately to be the most desireable, the flexibility and ease of switching from one particle to another is mandatory as is the minimization of time involved in this process and in-adjustment of the beam for use. Design should be such that repair can be rapidly effected e.g. the ion source, septum, deflector, electronics and regulation devices, etc. Multiple beam lines can provide efficient use of a machines capabilities and have become standard in all the new research installations. The question of fixed or variable energy can perhaps best be answered by noting that most of the newer installations are variable energy machines. The small extra cost and accompanying increased complexity is compensated by the greatly increased flexibility in use and by doing away with the bother of using beam-degraders (and their unwanted activation) in order to obtain a specific energy. However a fixed energy machine can have

advantages in a production context, if precise energy control is unnecessary.

Another consideration is external versus internal beam. An internal beam has the clear advantage of allowing utilization of near full current capability of the machine. However, this must be balanced against the increased engineering costs of target design and the fact that high current internal beams can only be used most effectively on metal targets or targets which have high radiolytic and chemical stability. External beams are considerably easier to work with and allow much greater flexibility in choice of target design and target. Research is most effectively done if external beams are available. Production of radionuclides may be more effectively done with internal beams but this depends on which nuclides are needed.

External beams should have an easily controllable shape and size. A cylindrical shape is usually best, optimally variable from 3-5 mm to 80 mm in diameter. The beam should have uniform density, and collimation should not result in appreciable loss of beam. Transfer efficiency to remote targets should be high, and current and beam density measurements should be easy to obtain. Targets and target materials should be thermally stable, chemically unreactive and radiolytically stable. External beams allow the facile bombardment of gases, liquids and solids. A vertical beam would be especially useful in the bombardment of liquids and corrosive solids because of facilitation of engineering design problems and window stability. The nuclide products which are frequently near carrier free should not readily be adsorbed on target surfaces.

Consideration of optimum beam characteristics however, must be considered in light of the machine application. A broad spectrum nuclide production program taking into consideration all parameters ranging from the nuclides required, production in large quantity, and in high radio-

nuclidic purity cannot be accomplished with a machine of high beam current but low particle energy. On the other hand, if the program requires production of a limited number of nuclides in large quantity especially if one is considering large scale use of nitrogen-13, oxygen-15, carbon-11 and fluorine-18 it would indeed be ill advised to invest in a high current, high energy machine.

A consideration of carbon-11, fluorine-18 and iodine-123 can be illustrative of this point. Tables VI and VII give the saturation activity values for a high current beam on target, for a number of particle energies incident on target, for carbon-11 [11] and fluorine-18 [12] respectively. Similar illustrations can be made using the $^{10}B(d,n)^{11}C$ and $^{11}B(p,n)^{11}C$ and $^{16}O(^{3}He,p)^{18}F$ [$^{16}O(^{4}He,pn)^{18}F$] reactions, however, highly accurate excitation functions over a broad range of energies are not given in the literature for e.g. the $^{10}B(d,n)^{11}C$ reaction. As is evident from the table a machine allowing for 9 MeV deuterons on target and 12 MeV protons on target would be a good comprimise for size (cost) versus capability optimum. Translating this by including window thickness and considering that the maximum deuteron energy available in a multiparticle cyclotron is roughly 0.5 x the proton energy for any given machine, to what sort of machine one might want for routine production for daily clinical use, one would argue for a multiparticle machine in the external proton beam energy range of 14-20 MeV. Requirements are roughly comparable for the other important carbon-11 and fluorine-18 producing reactions. Integrated into these considerations must be an understanding of the difference between producing 1 curie of $^{11}CO_2$ or 100 mCi of $^{18}F^-$ (e.g.) for direct use, both of these forms being readily available at EOB, and producing a complex compound for delivery; one to three half lives after EOB especially if the yields of these compounds are not high.

Turning to another aspect let us consider iodine-123. There is now

a general consensus that production of iodine-123 is best done by using either tellurium enriched to 90% or better in tellurium-124 [13] in the $^{124}Te(p,2n)^{123}I$ reaction, or by using the $^{127}I(p,5n)^{123}Xe \rightarrow ^{123}I$ reaction (or the near equivalent $^{127}I(d,6n)^{123}Xe \rightarrow ^{123}I$ reaction). There is also little "remaining" doubt that the ^{123}I from the p,5n is radionuclidially the most pure (completely aside from the auxilliary advantages of going through the ^{123}Xe parent). The choice is based on the cyclotron available. The p,2n can be effectively carried out to give high yield if the available proton energy is 28 MeV or a bit higher. The p,5n reaction requires a machine of at least 60-70 MeV capability.

The question of optimum size is clearly complex and as yet no definitive answer exists, however, directions for proposed application are clearly evident.

Cyclotrons - The Future

Future machines should be considered in the context of what their application will be and whether or not alternate modes for desired nuclide production are possible.

A number of possibilities exist for alternate accelerators. Linacs of varying types should be considered especially if only one particle such as protons or electrons is required. Proton linacs are already being used for nuclide production (cf table III). If such devices at more modest energies could be designed at low cost they might become a viable alternative to the medical cyclotron. Electron linacs, where one uses a target, usually tungsten, to produce bremstrahlung,hence allowing the use of photonuclear reactions, has been nicely addressed by Welch [14]. The use of Van de Graaff accelerators is not promising because of limitations on current and particle energy.

In the cyclotron area new concepts to increase flexibility and to

reduce cost,include the self shielding concept incorporated in the JSW "Baby"cyclotron this in principle obviating the need for heavy shielding walls (in practice, certainly reducing shielding requirements). Another possibility is that the septum deflector requirement could be replaced with field manipulation giving a lower quality external beam, but one which would be more than adequate in the medical context.

The all-in-one concept for hospital use is one that has gained support in the last several years, an interest stimulated in good part by the remarkable advances in sophistication in imaging instrumentation, especially positron tomography. Such a cyclotron would be a complete factory in one unit requiring mainly operator manipulation in order to produce needed short lived isotopes for clinical application and reducing or eliminating the need for elaborate post bombardment processing facilities requiring highly skilled personnel.

However the need for more reliable more flexible and less expensive machines is still with us. The needs of a hospital versus those of a University Medical Center or large research center are clearly different. Regrettably no extensive study of all the factors involved including the economic factors has to date been carried out. It is this authors contention that there is no one size or set of specifications for a cyclotron that will satisfy all requirements. The use of cyclotrons for nuclear medicine is an evolving art. The numbers of machines listed in table V is indeed deceptive since it does not accurately reflect the complex physical, biological and medical research commitment as it currently stands and what is required for the future in order to allow accelerator produced nuclides to be an important a factor in the delivery of health care to the public.

I do not share the uneasiness if not malaise apparent in some

quarters for the future of nuclear medicine because of the advent of the "CAT" scanner and ultrasound diagnostic instruments. Nuclear medicine had made great strides in the area of morphology study and function study but the application of nuclear medical techniques to more sophisticated approaches to function and metabolism is really in its infancy. One should only consider that the great majority of the cyclotron facilities listed in Table V were installed or evolved programs in support of nuclear medicine after 1970. One of the many exciting possibilities for the future is the "packaged" cyclotron installation for nuclear medicine at a "reasonable" cost. A possibility brought about by a marriage of research in physics, chemistry and medicine.

The true value of cyclotrons and other accelerators in basic research in biology and medicine has already been demonstrated and clinical application in some areas is already routine. I am indeed optimistic for the future of the Inter-Multi Disciplinary Field of Nuclear Medicine and Medical Accelerators.

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Table I. Medical Cyclotrons

Company	Cyclotron Corp.				CGR-MeV	Scandatronix
Particle \ Type	CS15	CS22	CS30	CV28[b]	CGR 520[b]	MC30[b]
Protons[a]	15	22	26	24	24	30
Deuterons	8	12	15	14	12.5	15
Helium-3	20	30	38	36	33	40
Helium-4	16	24	30	28	25	30

a Maximum particle energies in MeV

b Variable energy

Table II. Medical Cyclotrons

Company	CGR-MeV		Japan Steel	BNL
Type / Particle	CGR-680[b]	CGR930[b]	"Baby"	152Cm[b,c]
Protons[a]	38	90	9.4	36
Deuterons	25	53	4.7	24
Helium-3	—	—	12.5	65
Helium-4	50	110	9.4	46

a Maximum particle energies in MeV

b Variable energy

c The Brookhaven National Laboratory Cyclotron is included in this listing for comparison purposes only. It is a machine converted from research in physics and chemistry to research in nuclear medicine and chemistry.

Table III. Large High Current Accelerators-Symbiotic Relationship

Facility	Type	Particle	Current
1. BNL Injection LINAC for AGS; Facility BLIP[a]	LINAC	200 MeV protons	150 μA(200 μA)[f]
2. LAMPF[b]	LINAC	800 MeV protons	100 μA(1000 μA)
3. TRIUMF[c,d]	Cyclotron	500 MeV protons	100 μA (300 μA)
4. SIN Isochronous Ring[d,e]	Cyclotron	590 MeV protons	60 μA (100 μA)

a BLIP - Brookhaven Linac Isotope Production

b LAMPF - Los Alamos Meson Production Facility

c TRIUMF - Tri University Meson Facility, Vancouver

d Proposed use, not yet on stream.

e SIN - Schweizerisches Institut fur Nuklearforschung

f Design intensity is given in brackets.

Table IV. World Distribution of Cyclotrons Used in Nuclear Medicine Research and Application

Location \ Function	Dedicated[a]	Established Program[b]	Dedicated-Private[c]
1. North America	7	9	5
2. South America	-	3	-
3. Europe	7	20	1
4. Africa & Middle East	1	1	-
5. Asia (Japan)	3	2	1
TOTAL:	18	35	7

[a] A dedicated machine is one which is either wholly or primarily used in a nuclear medicine program for research and application.

[b] Machines listed under established programs are those which have as part of their overall use an established program in nuclide work for nuclear medicine purposes and may also be part of an ongoing onsite program in nuclear medicine. (Some machines have undoubtedly been inadvertently left out of this category because of the difficulty of identifying all programs in this category. The author would welcome information for any source on oversights).

[c] Included here are machines owned wholly by private companies or machines which were purchased and are supported in the main by private funds.

Table V. *Location of Cyclotrons Used for Nuclear Medicine Purposes*

North America		Type	Use[a]
1.	Washington University Medical School St. Louis, USA	Allis Chalmers	D
2.	Washington University Medical School St. Louis, USA	CS-15[b]	D
3.	Massachusetts General Hospital Boston, USA	Allis Chalmers	D
4.	New England Nuclear Boston, USA	CS22	D-P
5.	New England Nuclear Boston, USA	CS30[b]	D-P
6.	Sloan Kettering Institute New York, USA	CS15	D
7.	Brookhaven National Laboratory Upton, USA	152 cm[c]	D
8.	Medi Physics South Plainfield, USA	CS22	D-P
9.	Oak Ridge National Laboratory Oak Ridge, USA[b]	193 cm[c]	E-P
10.	Franklin McLean Chicago, USA (Prev. Argonne Cancer Hosp.)	CS-15	D
11.	Indiania University Medical Center Indianapolis, USA	660 cm[c]	E-P
12.	NASA-Lewis Research Center Cleveland, USA	175 cm[c]	E-P
13.	Argonne National Laboratory	152 cm[c]	E-P
14.	Mt. Sinai Hospital Miami Beach, USA	CS30	D-P
15.	University of California Los Angeles Medical School Los Angeles, USA	CS22	D
16.	Medi Physics Berkeley, USA	CS22	D-P

(continued)

North America	Type	Use[a]
17. University of California Berkeley, USA	——[c]	E-P
18. University of California Davis, USA	193Cm[c]	E-P
19. University of Washington Seattle, USA	——[c]	E-P
20. McMasters University Toronto, Canada	——[c]	E-P
21. McGill University Montreal, Canada	——[c]	E-P

South America	Type	Use
22. Institute of Nuclear Energy Rio di Janeiro, Brazil	CV-28	E-P
23. Institute of Atomic Energy Sao Paulo, Brazil	CV-28[b]	E-P
24. National Comm. Atomic Energy Buenos Aires, Argentina	——[c]	E-P

Europe	Type	Use
25. Hammersmith Hospital MRC London, U.K.	MRC[c]	D
26. Radiochemical Center Amersham, U.K.	140Cm[c]	E-P
27. Atomic Energy Establishment Harwell, U.K.	178Cm[c]	E-P
28. Western General Hospital MRC Edinborough, U.K.	CS-30[b]	D
29. Service Hospitalier Frédéric Joliot Orsay, France	CGR-520	D

(continued)

Europe	Type	Use
30. National Center of Scientific Research Orléans, France	CGR-580	E-P
31. Institute of Nuclear Sciences Grenoble, France	212 cm[c]	E-P
32. University of Louvain La Neuve Louvain La Neuve, Belgium	CGR-930 S	E-P
33. University of Liege Liege, Belgium	CGR-520	E-P
34. University of Ghent Ghent, Belgium	CGR-520[b]	E-P
35. University of Gronningen Groningen, The Netherlands	Philips-280 cm[c]	E-P
36. IKO Institute Amsterdam, The Netherlands	____[c]	E-P
37. Philips-Duphar Petten, The Netherlands	Philips-140 cm[c]	D-P
38. University of Eindhoven Eindhoven, The Netherlands	Philips-130 cm[c]	E-P
39. Central Hospital of Tampere Tampere, Finland	Russian(∿110 cm)[c]	D
40. University of Uppsala Uppsala, Sweden	230 cm	E-P
41. Scandatronix Uppsala, Sweden	MC-20	(?)
42. Medical School Hannover BRD	MC30[b]	D
43. Medical School Essen BRD	CV28[b]	D
44. Kernforschungsanlage Jülich Jülich, BRD	CV28	E-P
45. Kernforschungsanlage Jülich, BRD	330 cm[c]	E-P
46. Cancer Research Institute Heidelberg, BRD	AEG-109Cm[c]	D

(continued)

Europe	Type	Use
47. Kernforschungsgentrum Karlsruhe Karlsruhe, BRD	225 cm[c]	E-P
48. Institute for Reactor Research-SIN Würenlingen, Switzerland	Philips-250 cm	E-P
49. University of Milan Milan, Italy	166 cm[c]	E-P
50. University of Zagreb Zagreb, Yugoslavia	____[c]	E-P
51. Nuclear Research Institute Rez, Czechoslavakia	____[c]	E-P
52. Nuclear Research Institute Rossendorf DDR	____[c]	E-P
53. Institute of Physics & Power Engineering Obninsk, USSR	(∿110 cm)[c]	E-P

Africa and Middle East	Type	Use
54. King Faisal Hospital Riyadh, Saudia Arabia	CS-30[b]	D
55. Council for Scientific and Ind. Research Pretoria, Republic of South Africa	110 cm[c]	E-P

Asia	Type	Use
56. Institute for Physical Research Saitama, Japan	(∿160 cm)[c]	E-P
57. University of Tokyo Tokyo, Japan	CS30	D
58. Nihon Medi Physics Hyogo, Japan	CS30	D-P
59. Tokyo Chest Hospital Tokyo, Japan	JSW-Baby[b]	D

(continued)

60.	National Institute of Radiological Sciences Chiba, Japan	CGR-930	D
61.	Tohoku University	CGR-520(?)[b]	E-P

a- cf notes a and b table IV. An estimate of actual use is not given for those machines listed under E.P. It can vary from 2-3% of beam time to 50% of beam time or more. Thus a cyclotron installation marked E.P. may have a many faceted nuclear medicine program in operation (e.g. Jülich) while others may have a regular medical nuclide program but no extensive nuclear medicine programs connected with the nuclide production.

b- Machines with the notation b are in the process of construction, or installation and testing.

c- If a machine was not wholly produced by a commercial manufacturer or if specifications could not be found, it was listed with the designation c. These machines vary widely in particle capability, external beam current if any, and age. The diameter of the pole face is given (where available) in cm as a rough indication of the capability of the machine. For comparison a CS-30 has a pole face diameter of 96.5 cm.

Table VI. Thick Target Yields Fluorine-18 $^{20}Ne(d,\alpha)^{18}F$

Energy Incident on Target	Saturation Value Ci/50 μA
3	0.200
6	1.4
9	2.9
12	3.9
15	4.4

Table VII. Thick Target Yields Carbon-11 $^{14}N(p,\alpha)^{11}C$

Energy Incident on Target	Ci/100 μA Saturation
6	0.330
9	3.7
12	6.6
15	10.6