REVIEW OF CYCLOTRONS USED IN THE PRODUCTION OF RADIO-ISOTOPES FOR BIOMEDICAL APPLICATIONS

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Abstract

Cyclotrons are the primary tool for producing the shorter-lived proton-rich radio-isotopes currently used in the biosciences. Although the primary use of the cyclotron produced short-lived radio-isotopes is in PET/CT and SPECT diagnostic medical procedures, cyclotrons are also producing longer-lived isotopes for therapeutic procedures. Commercial suppliers are responding by providing a range of cyclotrons in the energy range of 3 to 70 MeV. The cyclotrons generally have multiple beams servicing multiple targets. This paper provides a comparison of some of the capabilities of the various current cyclotrons. The use of nuclear medicine and the number of cyclotrons providing the needed isotopes is increasing. In the future it is expected that there will be a new generation of small 'table top' cyclotrons providing patient doses on demand.

INTRODUCTION

Cyclotrons have become the tool of choice for producing the short-lived. proton-rich radio-isotopes used in biomedical applications [1]. Industry has responded with a variety of cyclotrons to address the particular needs of different users groups. Most of these machines have been installed in hospitals, institutes for academic research, and commercial facilities specializing in producing and selling of radio-isotopes. Cyclotrons for biomedical radionuclide production are generally compact, accelerate light ions (proton, deuteron or helium) and are primarily used to produce short-lived, proton-rich radio-nuclides. The main use of these unstable isotopes is for diagnostics and therapy in biomedicine. Other fields using radio nuclides as tracers include agriculture (bio-kinetics in plants and soil), biology (bio-chemical and toxicological studies), ecology (pollution, environmental impact, and ecology studies), Geology (migration of elements in soils and waters) and pharmacology (metabolic studies). The use and need of radio-active isotopes for biomedical applications continues to increase worldwide. However, the list of radio nuclides and the applications have not changed significantly over the past 20 years [2].

Five years after demonstrating the first cyclotron in 1931 [3], G. Lawrence was producing phosphous-32 with by an accelerator and for injection into a patient with chronic leukaemia. Other isotopes generated by his cyclotron also had important applications in medicine. However, his vision to develop a radiopharmaceutical industry at the Radiation Laboratory to help sustain accelerator physics in the late 1930s was an idea ahead of its time. In 1941 the first cyclotron dedicated to the production of radioisotopes was installed at Washington University, St Louis and was used to produce isotopes of phosphorus, iron, arsenic and sulphur. In the mid 1950s a group at Hammersmith Hospital in London put into operation a cyclotron wholly dedicated to radionuclide production. Scanditronix was founded in 1961 as a private company to commercialize cyclotrons for use in the medical field.

Radioactive isotopes have both diagnostic and therapeutic applications in Nuclear Medicine. PET (positron emission tomography), PET/CT and SPECT (single photon emission computed tomography) are the main diagnostic procedures in nuclear medicine. Radioactive isotopes for biomedical applications are produced in reactors and with accelerators. Of particular importance for PET are the short-lived positron emitters, 11C, 13N, 15O and 18F. Carbon, nitrogen and oxygen represent basic constituents of organic matter and this characteristic permits the labelling of a great variety of radio-pharmaceuticals. SPECT uses medium-lived radio-nuclides that are single photon emitters. This is a technique in which a gamma camera rotates around the patient taking ‘pictures’ which a computer uses to form a cross-sectional tomographic image. Therapeutic applications prefer to use radionuclides that have a high linear-energy transfer associated with their decay products and that can be chemically attached to a biologically active molecule which preferentially attaches to a tumour site. Table 1 contains a list of the main radioisotopes produced by cyclotrons along with some of the reactions that are used to produce the radioisotopes. The production yields come from a variety of sources but primarily from a review article by Ruth et al, vary considerably depending on target design and the chemical form of the radioisotope molecule that is being used for measured the yields and in some cases from calculations [4].

The principle advantage of accelerator produced radioisotopes is the high specific activity (SA) that can be obtained via the nuclear reactions that produce an isotope that is chemically different from the target element. Another significant advantage is the smaller amount of radioactive waste generated in particle reactions compared to reactor produced radioactive isotopes. (SA is a measure of the number of radioactive decays from an isotope of interest per unit weight of an irradiated sample). Most of the reactions used are of the form (p,n), (p,2n), (p,), (p,xn) and to a lesser extent reactions involving D, 3He and 4He as the projectile. Measured cross sections for many of these reactions along with references for the measurements can be found in an IAEA report titled “Cyclotron produced radio-nuclides: Physical Characteristics and production methods” [5]. For proton/deuteron acceleration the negative ion is preferred in order to reduce activation around the cyclotron whereas for helium acceleration, the positive ions must be accelerated.

In 2005, an IAEA report estimated that, worldwide, there were about 350 cyclotrons that were primarily used

for the production of radio-nuclides [6]. Nearly 50% of these were in the 10-20 MeV energy range and about 75% of the cyclotrons were being used to produce ^{18}F for FDG.

Medical Isotope	Life-time $T_{1/2}$	Use	Nuclear Reaction	Target Abun-dance (%)	Energy Range (MeV)	Production Yield (mCi @ sat)	Typical Dose (mCi)
^{11}C	20.4m	PET	$^{11}B(p,n)$	80.3	8 - 20	40/μA	
^{11}C	20.4m	PET	$^{14}N(p,\alpha)$	99.6	12	100/μA	
^{11}C	20.4m	PET	$^{10}B(d,n)$	19.7	7	10/μA	
^{13}N	9.96m	PET	$^{13}C(p,n)$	1.1	5 - 10	115/μA	
^{13}N	9.96m	PET	$^{12}C(d,n)$	98.9	2 - 6	50/μA	
^{13}N	9.96m	PET	$^{16}O(p,\alpha)$	99.8	8 - 18	65/μA	
^{15}O	2m	PET	$^{15}N(p,n)$	0.36	10 - 15	47/μA	
^{15}O	2m	PET	$^{16}O(p,pn)$	99.8	>26	25/μA	
^{15}O	2m	PET	$^{14}N(d,n)$	99.6	8 - 6	27/μA	
^{18}F	109.8m	PET	$^{18}O(p,n)$	0.20	8 - 17	180/μA	5 - 20
^{18}F	109.8m	PET	$^{20}Ne(d,\alpha)$	90.5		82/μA	
^{64}Cu	12.7h	SPECT	$^{64}Ni(p,n)$	0.93	5 - 20	5/μA	
^{67}Cu	61.9h	SPECT	$^{68}Zn(p,2p)$	19.0	>40	0.02/μA	
^{67}Ga	78.3h	SPECT	$^{68}Zn(p,2n)$	19.0	20 – 40	4.5/μA	10
$^{82}Sr/^{82m}Rb$	25d/5m	PET	$^{85}Rb(p,4n)^{82}Sr$ Produces Rb	72.2	50 -70	0.18 /μAh	
^{99m}Tc	6h	SPECT	$^{100}Mo(p,2n)$	9.7	19	14/μAh	20
^{103}Pd	17.5d	Therapy	$^{103}Rh(p,n)$	100	10 -15	0.52/μAh	
^{111}In	67.2h	SPECT	$^{112}Cd(p,2n)$	24.1	18 - 30	6/μAh	3
^{123}I	13.2h	SPECT	$^{124}Xe(p,2n)^{123}Cs \rightarrow ^{123}Xe \rightarrow ^{123}I$	0.10	25 - 35	27/μAh	
^{123}I	13.2h	SPECT	$^{123}Te(d,2n)\ ^{123}I$	0.89	10 - 15	20/μAh	
^{124}I	4.1d	PET	$^{124}Te(p,n)$	4.7	10 - 18	0.1/μAh	
^{124}I	4.1d	PET	$^{124}Te(d,2n)$	4.7	>20	0.15/μAh	
^{186}Re	90.6h	Therapy /SPECT	$^{186}W(p,n)$	28.4	18		
^{201}Tl	73.5h	SPECT	$^{203}Tl(p,3n)^{201}Pb \rightarrow ^{201}Tl$	29.5	27 - 35	0.7/μAh	4
^{211}At	7.2h	Therapy	$^{209}Bi(\alpha,n)$	100	28	1/μAh	0.05-.01

Table 1: This table lists commonly used Cyclotron-Produced Isotopes, some of the possible nuclear reactions used to produce the isotopes, typical production yields and some key physical properties for the target and production isotopes.

It is convenient to categorize the cyclotrons into three broad (proton) energy ranges based on their primary function [7]. (For reasons based on efficiency and cost considerations, some facilities/manufacturers have chosen accelerators and reactions that also use deuterons and helium as a projectile.) These proton energy ranges are:

- Cyclotrons with proton energy less than 20 MeV are primarily used for producing positron emitting radio-nuclides. These PET isotopes tend to have short half-lives and the cyclotrons are located in regional centres/hospitals determined by the yield loss due to the delivery time from cyclotron to patient. Many of the cyclotrons have the capability of being shielded with close-packed steel and thereby reduce the need for the user to provide a heavily shielded bunker. The delivery time of the

radio-isotope, the patient dose requirement and the number of doses required per day lead to a cyclotron providing up to 50 μA per target. Many of the current cyclotrons have the capability of using multiple targets on each of two or more extracted beams.

- Cyclotrons with proton energies between 20 to 35 MeV are primarily used to produce many of the gamma-emitting radioisotopes (commonly used as imaging radio-isotopes for SPECT) as well as the production of several PET isotopes. The SPECT isotopes have medium half-lives and production generally takes place in dedicated facilities. The longer half-lives permit isotope delivery to more distant users and this leads to dedicated production facilities with high power targets and larger throughput.
- Cyclotron providing protons with energies of greater than 35 MeV are used in the production of a number of the isotopes used for radio-therapy. The primary need is for high current cyclotrons with currents in the 1mA range.

CYCLOTRON DESIGN CONSIDERATIONS

For the customer, the primary cyclotron specification is given by a minimum target yield for a given radio-isotope in the form of a particular molecule within a specified irradiation time. For the cyclotron designer, this user provided specification leads to a decision on the nuclear reaction to be used, the target material, the accelerated particle, the target chemistry, the energy and the cyclotron current on target. The accelerated particles must have adequate energy for the chosen nuclear reaction and adequate current to provide the required yields at that energy. Secondary specifications include; the number of targets, the target dimensions, the number of extracted beams (ports), shielding (facility) requirements, activation, and operating costs. Invariably, compromises are needed in order to arrive at a manageable facility budget.

Table 2 lists some of the important characteristics of the cyclotrons offered by many of the current industrial suppliers. The most common choice for the magnet is a conventional room temperature coil. Most manufacturers have the option of self-shielding for the cyclotrons when the maximum energy is less than 15 MeV. The customer must choose between vault (concrete) shielding versus the more expensive but compact arrangement of close packing steel around the cyclotron and targets. Earlier generations of cyclotrons accelerated mostly positive ions. Today, most of the cyclotrons accelerate H- and/or D- because of the ease of extracting multiple beams into targets that are separately shielded by steel or concrete. The choice of either internal or external targets impact the footprint of the system and also the radiation exposure for personnel servicing the cyclotron. For H- currents less than 200 μA an internal PIG ion source is the preferred cost effective choice. If H- currents greater than about 200 μA are required, then an external (Cusp) ion source is required. With internal targets and H+ acceleration, high currents are easily achievable with internal PIG type ion sources. Extraction of intense H+ beams invariably lead to severe component activation in the cyclotron.

CYCLOTRON SUPPLIERS

Commercial companies have responded to the varied user specifications with a number of basic cyclotrons with optional add-ons in an attempt to satisfy each particular need and budget. What follows is a list of several companies, arranged alphabetically and their key product specifications. A short comparative summary can be found in Table 2.

Advanced Cyclotron Systems Inc (ASCI)

ACSI fabricates and sells 14 MeV, 19 MeV (fixed and variable energy) 24 MeV and 30 MeV cyclotrons. Simultaneous dual extraction is available in each of the models. The 14 MeV cyclotrons are available in self- shielded and unshielded configurations. A new product line is the TR24 providing high-current, variable-energy extracted proton-beams in the energy range 15 to 24 MeV to partly bridge the energy gap between their 19 and 30 MeV models. The TR30 offers variable-energy extraction (15-30 MeV) with currents exceeding 1,200 A and simultaneous dual beams. The TR30 (TR30/15) can also be configured to provide dual-particle acceleration.

Advanced Biomarkers Technology (ABT)

ABT has recently introduced a new low power, small 'table-top' cyclotron with internal targets. The company website notes that the accelerator has an on-off switch, a target selector switch and a beam current switch. The cyclotron accelerates H+ from an internal ion source. The accelerator has a fixed-energy of 7.5 MeV, a target volume of 15 micro-litres and is designed to provide single-patient doses (millicuries) of 18F and 11C. At 1 A and 7.5MeV, 1 millicurie/minute of 18F is produced. Currents are in the range of 1 to 5 A. With shielding the total weight of the accelerator is 10.8 ton and only occupies about 2 square metres.

Best Cyclotrons Systems Inc (BSCI)

BSCI is bringing three new cyclotron models to market [8]. They are fabricating a fixed-energy 14 MeV H- cyclotron with internal ion source, having a total extracted current of up to 100 μA into four external beams. Their 35 MeV cyclotron has an external CUSP type ion source, two simultaneous extracted beams with energy variable from 15-35 MeV and an advertised maximum extracted current greater than 1.5 mA. A 70 MeV model accelerates H- from an external ion source, has simultaneous variable-energy extraction from 35-70 MeV into two external beam lines.

China Institute of Atomic Energy (CIAE)

CIAE is presenting, at this conference, details of their plans for 14 MeV and 70 MeV cyclotrons for the production of medical isotopes [9, 10, 11]. Both have external ion H-/D- ion sources. The CYCIAE-70 provides 700 A of extracted H+ at 35 - 70 MeV and 40 A of D+ at 18 - 33 MeV. The CYCIAE-14 is designed to provide two dual extraction ports servicing 4 different targets. It is a fixed-energy cyclotron providing up to 400 A at 14 MeV.

Efremov (NIIEFA)

The Efremov Institute supplies a cyclotron that accelerates negative hydrogen and deuterium ions, in a vertical plane, to energies of 18 and 9 MeV, respectively. The cyclotron uses a CUSP type external ion source and provides extracted currents of H/D at 100/50 A [12].

EuroMeV

EuroMeV offers the ISOTRACE superconducting cyclotron (based on the OSCAR-12 initially developed by Oxford Instruments). This cyclotron provides an extracted beam current up to 100 A at a fixed energy of 12 MeV. The cyclotron weighs only 3.8 tonnes and has a total operating power consumption of 40 kW.

GE Healthcare

GE Healthcare has two cyclotron products for PET, namely, MINItrace and PETtrace. MINItrace accelerates H- in a vertical oriented cyclotron that provides 50 A at a fixed-energy of 9.6 MeV. The PETtrace also accelerates in a vertical plane either H- /D- up to a fixed-energy of 16.5/8.6 MeV with extracted currents of 100/65 A. The MINItrace cyclotron features integrated shielding and fully automated operation during start-up, tuning and operation.

IBA

IBA markets cyclotrons at 3 (D), 10/5 (H/D), 11 (H), 18/9 (H/D), 30 (H) and 70/35 (H/D) MeV. Cyclone 3D is described at this conference [13]. Cyclone 3D was originally marketed to address the need for 15O in the early 90s. The original 4 pole geometry has been replaced by three poles to provide additional vertical focussing. Cyclone 10/5 is a fixed energy cyclotron with four extraction ports. Cyclone 11 is a new product line that features a fixed-energy, self-shielded cyclotron that accelerates H- up to 11 MeV [14]. The design is based on the Cyclone 10/5 cyclotron. Cyclone 18/9 is a fixed-energy cyclotron that accelerates H- up to 18 MeV and D- up to 9 MeV. Cyclone 18 Twin is a fixed-energy cyclotron that accelerate H- up to 18 MeV and that improves uptime and reliability by using two independent ion sources. Cyclone 30 is a fixed-energy cyclotron that accelerates H- up to 30 MeV and can extract two independent beams. Cyclone 30 XP is another new product line that accelerates proton, deuteron and alpha beams up to 30 MeV. Protons and deuterons are accelerated as negative ions and extracted by stripping and at variable energy. The alpha beam is accelerated as He++ and extracted with an electrostatic deflector [15]. Cyclone 70 is a multi-particle, fixed-energy cyclotron that accelerates H- up to 70 MeV as well accelerating alpha beams. The specifications for the Cyclone 70 installed in Arronax had the H- at variable energy from 35-70 MeV with a maximum current of 750 A, D- from 17-25 MeV at 50 A, He++ at 70 MeV and A and H2+ at a fixed 35 MeV and maximum current of 50 A.

KIRAMS

The Cyclotron Application Laboratory (KIRAMS) has developed two cyclotrons for radioisotope production. The KIRAMS-30 accelerates H- from an external ion source up to 30 MeV. The cyclotron extracts protons of up to 500 A into through two ports at energies from 15 – 30 MeV [16]. The KIRAMS-13 accelerates H- from an internal PIG ion source up to 13 MeV and extracts currents of up to 80 A through two ports.

Siemens

Siemens markets the Eclipse brand cyclotrons which were initially developed by CTI and sold under the RDS label. The Eclipse cyclotrons accelerate H- up to a fixed energy of 11 MeV. The Eclipse HP provides 60 A into each of two beam lines and onto a carousel that holds up to 2 targets whereas the RD provides 40 A on a carousel that holds up to 8 targets in each of two beam lines. The Eclipse ST is a self-shielded HP.

Sumitomo

Sumitomo has built a series of cyclotrons under the names HM-7, HM-10, HM-12S, HM-12 and HM-18. Each accelerates both H- and D-. The HM-7 is a fixed energy cyclotron that has a self-shielding option and provides H/D at an energy of 7/3.5 MeV. The HM-10 is also a fixed-energy cyclotron that accelerates H-/D- to 9.6/4.8 MeV, can be equipped with 5 targets and has a self shielding option. The HM-12 cyclotron accelerates H-/D- up to 12/6 MeV, has two extraction ports that can each be equipped with up to 4 targets. The HM-12S is the shelf-shielded version of the HM-12. The HM-18 cyclotron is also a fixed energy cyclotron accelerating H-/D- up to 18/10 MeV, has two extraction ports each accommodating four targets. All of the Sumitomo cyclotrons use internal PIG ion sources.

Others

There are numerous facilities still using cyclotrons that were manufactured by companies that no longer offer cyclotrons for sale. These cyclotrons include products originally manufactured and sold by Scanditronix (MC series cyclotrons), The Cyclotron Corporation (TCC series cyclotrons), Japan Steel Works (JSW/BC series cyclotrons), and Computer Technology and Imaging Inc (CTI with their CTI-RDS, Radio Isotope Delivery System cyclotrons).

Company Name	Cyclotron Model	Particles	Energy (MeV)	Beam Current (μA)	Ion Source Type	Peak Field (T)	Hill Valley ratio	RF Freq. (MHz)	Plane Of Accel	Cyc. Weight (t)	Shield Weight (t)	Power (kW)
ACSI	TR14	H-	14	>100	Cusp	2.1		74	V	22	40	60
ACSI	TR19/(9)	H-/(D-)	19/(9)	>300/100	Cusp	2.1		74/37	V	22		65
ACSI	TR24	H-	24	>300	Cusp	2.1	4	83.5	H or V	84		80
ACSI	TR30/(15)	H-/(D-)	30/(15)	1500/400	Cusp	1.9			H	56		150
ABT	TableTop	H+	7.5	5	PIG	1.2		72	H	3.2	7.6	10
Best	BSCI 14p	H-	14	100	PIG			73	H	14		60
Best	BCSI 35p	H-	15-35	1500	Cusp			70	H	55		280
Best	BSCI 70p	H-	70	800	Cusp	1.6		58	H	195		400
CIAE	CYCCIAE14	H-	14	400	Cusp							
CIAE	CYCCIAE70	H-	70	750	Cusp							
NIIEFA	CC-18/9	H-/D-	18/9	100/50	Cusp			38.2	V	20		60
EUROMEV	Isotrace	H-	12	100	Cusp	2.36	(SC)	108	V	3.8		40
GE	MINItrace	H-	9.6	>50	PIG	2.2	~2	101	V	9	40	35
GE	PETtrace	H-/D-	16.5/8.6	>100/65	PIG	1.9	~1	27.2	V	22	47	70
IBA	Cyclone 3	D+	3.8	60	PIG	1.8	1	14	H	5		14
IBA	Cyclone 10/5	H-/D-	10/5	>100/35	PIG	1.9	~5	42	H	12	40	35
IBA	Cyclone 11	H+	11	120	PIG	1.9	~5	42	H	13	52	35
IBA	Cyclone 18/9	H-/D-	18/9	150/40	PIG	1.9	~5	42	H	25		50
IBA	Cyclone 30	H-/(D-)	30 /(15)	1500/?	Cusp	1.7	14		H	50		180
IBA	Cyclone 70	H-/α										
IBA	Cyclone 70 (Arronax)	H-/D- /H_2^+/He^{++}	30-70/15-35 /17.5/70	2x350/50 /50/35		1.7	14	66/30	H	125		350
KIRAMS	KIRAMS-30	H-	15-30	500	Cusp	1.9	8	64	H			
KIRAMS	Kotron-13	H+	40372	100	PIG	2	2	77.3	H	20	80	18?
Siemens	Eclipse RD	H-	11	2x40	PIG	1.9	~13		H	11	39	35
Siemens	Eclipse HP/ST	H-	11	2x60	PIG	1.9	~13	72				35
Sumitomo	HM-7	H-/D-	7.5/3.8						V		30	
Sumitomo	HM-10	H-/D-	9.6/4.8						V		52	
Sumitomo	HM-12/S	H-/D-	12/6	>60/30	PIG	2	~5	45	V	11	56	45
Sumitomo	HM-18	H-/D-	18/10	>90/50	PIG	2	~4	45	H	24	86	55

Table 2: This table lists current manufactures of cyclotrons used in radioisotope production for medical applications, their cyclotron models, and compares some key cyclotron specifications.

FUTURE

The total number of cyclotrons producing radionuclides is gradually growing to meet the expanding needs of nuclear medicine. In some regions, physicians now require a PET/CT scan before setting up a treatment protocol for certain diseases. This growth that has been observed in the past decade is expected to continue. New cyclotron models adapted to current needs are being designed.

The recent shortage of reactor produced 99- molybdenum used in 99m-techecium generators, has revived interest in the possibility of accelerator production. 99Mo is currently produced in older research reactors using highly enriched uranium (HEU). Both the security issue surrounding the safe storage of the HEU and also the expected lifespan of these older reactors has given urgency to pursuing alternate production methods. Approximately 80% of nuclear medicine procedures currently use 99mTc. The molybdenum isotope of mass 100 (100Mo) bombarded with ~15 - 19 MeV protons could be used to supply regional amounts of 99mTc directly through the (p, 2n) reaction [17, 18]. High-current cyclotrons are needed to meet the demand and to avoid the build-up of ground-state contamination, but the high-power target technology will be challenging.

The primary PET isotopes are 11C and 18F. In most cases the cyclotrons producing these isotopes run in batch mode producing 10s of curries per run. Patient doses on the other hand tend to be a factor of 1000 smaller; i.e.; 10s of millicurries. Hospital clinical users have expressed interest in small machines that provide single patient doses with the touch of a button. Visionaries are suggesting that we will soon have small 'table-top' accelerators with targets using micro-fluidics to carry out the chemistry on a 'chip', providing the radio-pharmaceuticals in patient doses prior to each procedure. One supplier (Advanced Biomarkers Technology, USA) has already developed a ‘table top’ cyclotron which promises to provide patient doses on demand.

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