

## HIGH INTENSITY CYCLOTRONS FOR RADIOISOTOPE PRODUCTION or THE COMEBACK OF THE POSITIVE IONS

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High current cyclotrons are needed for the production of radioisotopes for nuclear medicine or for radiotherapy. Another application of high current cyclotrons is the production of  $^{99m}\text{Tc}$ , either directly or as a fission product in a cyclotron-driven subcritical neutron source. The first cyclotrons for radioisotope production were classical, positive ions accelerators with E.S.D. extraction. The extracted beam intensity was limited to 100  $\mu\text{A}$  or less. Internal targets were used when higher currents were needed. The TCC model CP 42 was the first to introduce negative ions extraction, but still used an internal source. In 1987, IBA introduced the CYCLONE 30, featuring an innovative magnet design and an external ion source. Currents for such machines escalated over time to more than 1 mA extracted, with 2 or 3 mA in view. But, for very high intensities, positive ion acceleration still presents significant advantages, as illustrated by the CYCLONE 18+, operating daily at 2 mA continuously on an internal target. We describe a new method for the extraction of positive ions from a cyclotron without the need of a deflector or a similar device.

### 1. The needs for high beam currents

#### 1.1 *The production of the "classical" radioisotopes for nuclear medicine*

A significant fraction of the nuclear medicine diagnostic procedures (around 20%) uses radioisotopes produced by cyclotrons. Among these, Thallium 201, Iodine 123, Gallium 67 and Indium 111 are the most commonly used<sup>1</sup>. These isotopes are produced by radiopharmaceutical companies in cyclotron based production facilities, and are then distributed to the hospitals on a domestic or international scale. These isotopes are all produced with 23 to 32 MeV proton beams. Obviously, the production of large amounts of isotopes in a limited time requires high beam currents. Today, the beam currents used are limited more frequently by the target technology than by the cyclotron limits. With the exception of the  $^{123}\text{I}$ , which is today normally produced on a  $^{124}\text{Xe}$  gas target, the production is normally done on a thin metallic target of rare and expensive enriched isotope, deposited on a strongly water cooled base, normally made of copper, and placed at grazing incidence with respect to the beam (in order to enlarge the beam spot on the target surface and to reduce the target thickness). The heat transfer problems in the target assembly are critical, and have been the subject of many developments.

#### 1.2 *Radioisotopes for the radiotherapy of cancer*

In addition to the radioisotopes used for imaging and diagnosis in nuclear medicine, some radioisotopes are used for cancer therapy. Typical radioisotopes used for cancer therapy emit short range radiation, and have generally longer

half-life. This longer life, combined with the fact that the activities used for therapy are orders of magnitude higher than the activities used in diagnostic results in the fact that the beam currents needed for the production of these isotopes are also much higher and are generally expressed in milliamperes. In 1992 IBA was asked to develop a very high intensity, 18 MeV cyclotron for the production of the radioisotope  $^{103}\text{Pd}$ . This radioisotope is marketed, in small sealed sources, for the local treatment of prostate cancer by the company Theragenics in Atlanta (Ga). It can be produced by a (p,n) reaction on Rhodium, a good material for internal target. However, the reaction yield is low and large beam currents are needed to achieve the desired production levels.

#### 1.3 *The production of $^{99m}\text{Tc}$*

The most frequently used radioisotope for nuclear medicine,  $^{99m}\text{Tc}$ , is not produced today with cyclotrons, but is distributed as  $^{99}\text{Mo} \Rightarrow ^{99m}\text{Tc}$  generators. The preferred reaction for the production of  $^{99}\text{Mo}$  is the neutron induced fission of highly enriched uranium 235 targets in nuclear reactors. Most of the nuclear reactors used for this production are due, in the next years, for a major refurbishment or for decommissioning<sup>2</sup>. This problem of the future availability of nuclear reactors suitable for  $^{99}\text{Mo}$  production has prompted a renewed interest on alternative production methods.

Two alternative production methods, based on cyclotrons, are presently under development:

- The first method is based on the direct accelerator production of  $^{99m}\text{Tc}$  or  $^{99}\text{Mo}$ . For this production, three possible nuclear reactions have been investigated by M. Lagunas Solar and his team at U.C. Davis<sup>3</sup>. The technical

feasibility of these alternative production methods is being evaluated. Among others, there are questions regarding the specific activity of “instant Tc” and the separation chemistry of  $^{99}\text{Mo}$ , as well as licensing and distribution issues. Nevertheless, directly produced Tc could become, in the future, a product complementary to generator produced Tc. In this case, the most promising methods will require high intensity proton beams, in the 2 to 5 mA range, with energies around 25 MeV.

- The other method is the proton-driven fission neutron source for the production of fission  $^{99}\text{Mo}$  as proposed by IBA<sup>2</sup> and presented in another paper to this conference. The IBA proposal is based on a 150 MeV, 1.5 mA cyclotron driving a sub-critical intense neutron source, generating a thermal neutron flux of  $2.10^{14} \text{ n s}^{-1} \text{ cm}^{-2}$ , similar in intensity to these of the nuclear reactors used for the production of  $^{99}\text{Mo}$ .

## 2. The early positive ion cyclotrons for radioisotope production

The first cyclotrons produced commercially for the production of medical r.i. were “classical” isochronous cyclotrons, similar to their counterparts built for nuclear physics research, where the beam was extracted using an electrostatic deflector (ESD). The heat dissipation limits in the deflector septum limited the extracted beam power to a couple of kW, but higher intensities, typically several hundred  $\mu\text{A}$  were available on internal targets. The Cyclotron Corporation (TCC), based in Berkeley (Ca), produced between 1968 and 1984 in excess of 30 cyclotrons based on this technology

## 3. Introduction of negative ion technology: the CP42 from TCC

A major step was made in the early 80's when TCC proposed a 42 MeV cyclotron accelerating negative ions, produced by an internal P.I.G. source, and extracting by stripping. The ESD limit on extracted beam power was broken, and extracted beam currents exceeding  $200 \mu\text{A}$  up to 40 MeV were available. Thanks to the stripping extraction, variable energy extracted beams could be obtained in a fixed field, fixed frequency cyclotron. The simultaneous extraction of two beams was also, in principle possible. However, the use of an internal ion source meant a poor vacuum in the cyclotron accelerating tank, and significant amounts of beam were lost in the median plane by stripping on the residual gas. The need to deal with these large beam losses resulted in a quite complex design of the dees and of the R.F. systems, and resulted, at least in the beginning, in a lower reliability. This cyclotron was, initially, a commercial success. Six CP42's were ordered and built, even before the

prototype was fully tested. Delays in the commissioning of these cyclotrons contributed eventually to the bankruptcy of TCC. After this significant failure, negative ion technology was rejected by radioisotope producers for some time, in favor of more classical positive ion isochronous cyclotrons, like the MC40 model, made by the Scanditronix company in Upsala (Sw.). It should be noted, however, that the CP 42 are still operating today reliably and efficiently after an initially difficult startup and commissioning.

## 4. The Cyclone 30 and the modern, high current cyclotrons for radioisotopes production

The next major step in r.i. production cyclotrons was the introduction, in 1987, by IBA of the CYCLONE 30. The CYCLONE 30 introduced major improvements to the design of cyclotrons for r.i. production<sup>4</sup>. The first major improvement was the use of an external, multicusp ion source for the production of the negative ions. The ions are injected axially into the cyclotron magnet, so that the neutral gas released by the ion source can be pumped externally. The second major improvement was the introduction of the “deep valley” magnet design, combining the advantage of solid pole and separated sectors cyclotrons, and allowing to improve significantly the power efficiency of the accelerator. Typically, previous positive ion cyclotrons were using 300 kW of electricity to get 3 kW of extracted beam (1% power conversion efficiency), while the CYCLONE 30 required only 90 kW of electricity to produce 15 kW of extracted beam (16% power conversion efficiency). The CYCLONE 30 was the first cyclotron to demonstrate the possibility to have a beam loading exceeding 50 % of the total R.F. power. The CYCLONE 30 was so clearly superior that it made the previous design obsolete overnight. A total 16 CYCLONE 30 have been built and installed today by IBA.

The first CYCLONE 30's used a 2 mA external multicusp ion source made by IBA, and a 25 kW output R.F. final amplifier. The extracted beam intensity was  $500 \mu\text{A}$  (design value),  $350 \mu\text{A}$  guaranteed, with actual maximum currents varying from machine to machine between  $450 \mu\text{A}$  and  $600 \mu\text{A}$ . In order to reach higher currents, the latest CYCLONE 30 was equipped with a more powerful, 40 kW RF amplifier, and four, high pumping speed cryopumps. A new ion source, able to produce 7 mA of H<sup>+</sup> into a small emittance, was developed for IBA by A.E.A Technology in Culham (UK). The new source was tested on a cyclotron under tests in the IBA factory, and 1.2 mA of beam were accelerated and measured on a beam stop at 1.5 MeV. The lack of appropriate shielding prevented a high current test at full energy.

In the quest for still higher currents, A.E.A Technology has tested an improved multicusp ion source, able to produce 25 mA of H<sup>+</sup> into an emittance of  $0.16 \pi.\text{mm.mrad}$  (normalized). With this ion source, biased at a

somewhat higher voltage (45 kV instead of 30 kV used presently), extracted beam currents in excess of 3 mA should be available.

**5. Positive ion, multimilliampere cyclotrons for r.i. production: the Cyclone 18+**

The production of the <sup>103</sup>Pd isotope for cancer treatment requires a proton current in excess of one milliampere, at an energy of 15 to 18 MeV. An internal target being acceptable for this application, IBA designed a very high current positive ion, internal target cyclotron: the CYCLONE 18+. This cyclotron has demonstrated the possibility to provide continuous, unattended operation at 2 mA on target for one week irradiation's. An interesting feature of this cyclotron is the beam loading factor, reaching 80 % of the total R.F. power, with only 20 % being used to maintain the accelerating field. Also a 5 mA current was observed during factory tests on a beam dump at lower energy (the maximum current was actually limited by the melting of the beam dump, not by the ion source or space charge limits). Space charge calculations made for cyclotrons that do not require turn separation indicate that the intensity limit due to the axial component of the space charge forces is probably around 10 mA average beam current<sup>5</sup>. This applies not only to positive ion cyclotrons using an internal target but also to negative ion cyclotrons where the extraction is made by charge exchange.

**6. An innovative concept of auto-extraction or the rebirth of the positive ion technology**

The use of negative ion technology allows an extraction by means of stripping of the H<sup>-</sup> ions in a thin carbon foil, leading to an extraction efficiency close to 100%. This technology is therefore, up to now, the technology of choice for applications where high intensity beams must be extracted. However, the requirements on the vacuum quality are high and, to avoid electromagnetic dissociation, low magnetic fields must be used. The consequence is that high energy cyclotrons quickly become very large machines if negative ions are accelerated, which is nevertheless necessary if high intensity extracted beams are required. From this point of view, any technological innovation leading to the use of positive ion technology together with an extraction system allowing a nearly 100% extraction efficiency would represent an unquestionable improvement.

IBA is presently working on a totally new concept for the extraction of high currents of positive ions. This new concept, called the auto-extraction will provide close to 100% extraction efficiency without the need of extraction elements that could easily be damaged by high currents, like septa for electrostatic or magnetic extractors.

*6.1 Basic principles of the auto-extraction*

In an isochronous cyclotron, the average field increases with radius to compensate the relativistic mass increase of the accelerated particles. Close to the pole edge, it become impossible to maintain an isochronous radial field profile. The actual field falls below the ideal field, reaches a maximum, and starts to decrease. When the actual field starts departing from the ideal isochronous field, the accelerated particles start to lag with respect to the accelerating voltage on the dee. When the phase lag reaches 90°, the acceleration stops: this point represents the limit of acceleration. At an other (generally larger) radius, the field index, defined as  $N = R/B \text{ dB/dR}$ , reaches the value -1. This point, corresponding to the maximum of the product field times radius, is the limit of radial focusing. Past this point, the magnet is unable to hold the ions, and the ions escape the influence of the magnetic field. We call the radius where N reaches -1 the limit of self-extraction. If the gap is large, like in most existing cyclotrons, the radial fall of the field is quite gradual, and the limit of acceleration is found at a radius significantly smaller than the limit of self-extraction. Transporting the beam to the first limit to the second is the task of the extraction system, including generally an electrostatic deflector.

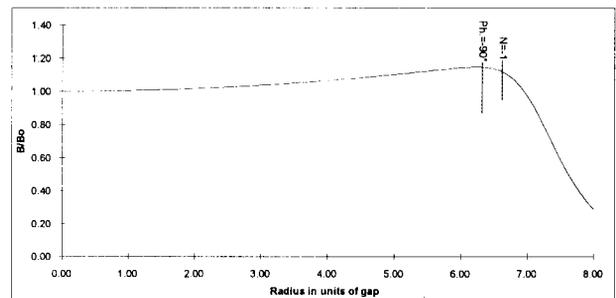


Figure 1: Magnetic field versus radius in cyclotron with R = 7 gaps

In a magnet with a smaller gap, the fall of the magnetic field close to the pole boundary is much sharper. As a result, the limit of acceleration falls much closer to the limit of self-extraction, and the extraction is much easier. This principle was a the center of the design of the 235 MeV cyclotron for protontherapy designed by IBA.

When the magnet gap at extraction becomes very small (like smaller than 20 times the radius gain per turn at extraction), the limit of self extraction is reached before the limit of acceleration, and the beam escapes spontaneously the magnetic field when the pole edge is reached.

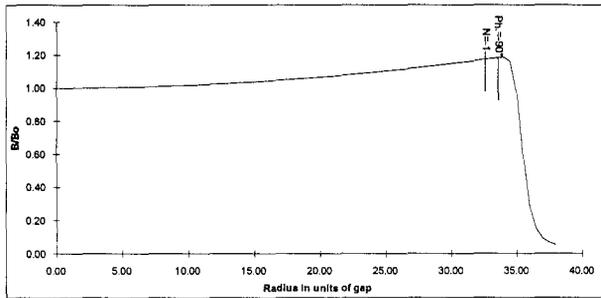


Figure 2: Magnetic field versus radius in cyclotron with  $R = 35$  gaps

### 6.2 Auto-extraction in the C235

It is interesting to note that this feature of self extraction is actually predicted by accelerated orbit simulations in the above mentioned cyclotron. Figure 3 features the tracking of 50 protons randomly distributed in the radial emittance of  $2\pi$  mm.mrad, in the magnetic field of the C 235 cyclotron, starting from a point on the equilibrium orbit at 230 MeV. All protons are found extracted, distributed all around the cyclotron, with an energy of 238 MeV. Clearly, the auto-extraction works, but in this form is not usable. It should be noted, however, that the C235 magnet was never designed for auto-extraction, and that its ability to self-extract is accidental. However, this calculation is useful because we plan to verify this crude first auto-extraction during the internal beam tests in the factory in the spring 1996.

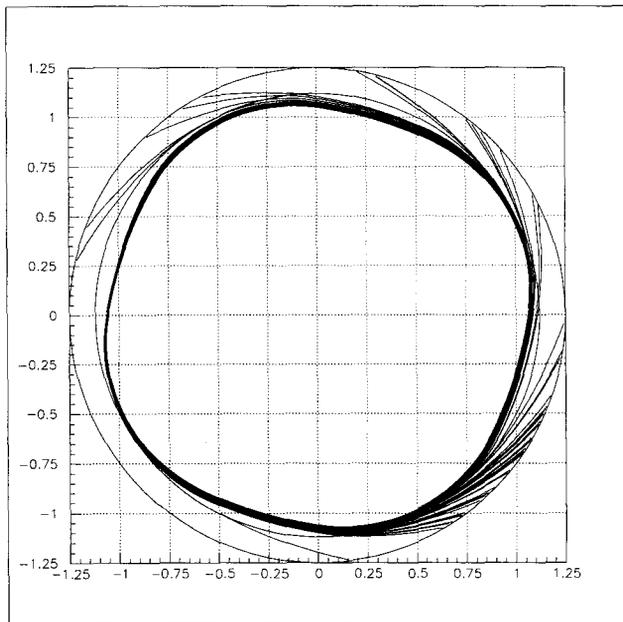


Figure 3: Tracking of self extracted protons in the C235 cyclotron

### 6.3 Orderly versus disorderly auto-extraction

The particles computer simulation made on the C235 indicate an extremely disorderly auto-extraction process, resulting in a non usable extracted beam. We have investigated if a more orderly auto-extraction was possible, in a cyclotron magnet designed for this purpose. To this effect, we have designed a low energy cyclotron, with four sectors. The magnetic field has been calculated using the TOSCA code using a realistic magnet design, and then has been isochronized. The magnet gap is 10 mm. The hill field is 2 Tesla. A first harmonic perturbation of 0.015 T has been introduced at large radii by a modification of two opposite sectors shape. At this radius, the radial betatronic frequency  $\nu_R$  is close to 1, so a  $\nu_R = 1$  resonance is excited at extraction. The dee voltage of 50.3 kV on two dees, or 25.15 kV on four dees results in a radial gain of 3 mm/turn at the pole radius of 350 mm. We track 50 protons, randomly distributed in the radial beam emittance of  $11.2\pi$  mm.mrad. This corresponds to a radial size of the beam of 3.3 mm. No phase spread has been introduced in this calculation. The particles are tracked from the start of the equilibrium orbit at 8 MeV, and followed until they escape the field.

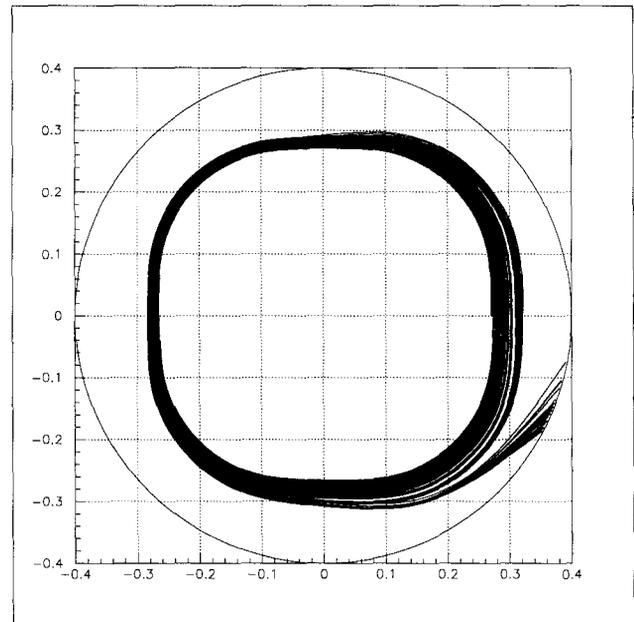


Figure 4: Tracking of self extracted protons in an optimized magnetic field

We see on figure 4 that, for an appropriate choice of dee voltage, we can obtain a very orderly auto-extraction of the beam. The extraction efficiency is 100%, and all the extracted beam seems usable. The radial divergence

observed corresponds to the well known radially defocusing effect of the magnet fringe field crossing, and could easily be corrected by a three iron bars gradient corrector. To understand better the conditions required to obtain an orderly auto-extraction, we have plotted (figure 5) the beam current profile at extraction, and we have calculated the field index at some specific points.

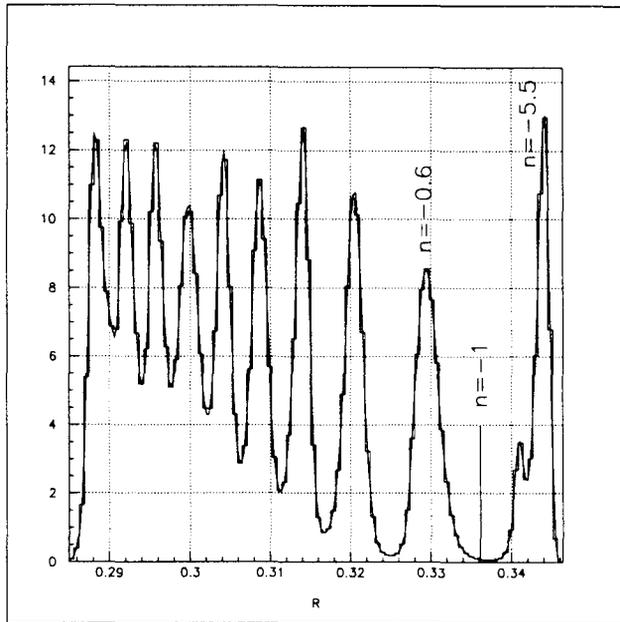


Figure 5: Radial density profile in the beam of figure 4

We see that the condition to be met to reach a nice extraction is to place the self-extraction limit,  $N = -1$ , in a zero current density between the last internal turn and the extracted beam. The  $v_R = 1$  resonance, excited at extraction by the first harmonic perturbation of the magnetic field, has increased the natural turn separation from 3 mm to 14 mm. As a result, the last internal turn sees a field index of  $-0.6$ , while the index reaches  $-5.5$  for the extracted beam.

The condition for a clean auto-extraction is, therefore, the possibility to generate, through a resonance or any other means, a turn separation similar to the size of the gap at extraction. The next question is: how small can the gap be at extraction? We believe that nothing prevents the gap to become almost as small as the axial beam size at that radius, leaving just enough margin to limit the pole irradiation particles in the tails of the axial distribution. The fact that, in a sector focused cyclotron, the flutter and, consequently,  $v_z$  increases with radius is favorable in this respect. The beam axial size is largest near the center, reducing the effect of the axial space charge forces in the region where their effect is maximum, and this size decreases slowly with radius as  $v_z$  increases. It will be generally favorable to locate slits limiting the axial emittance

near the center, before the ions have reached the threshold energies for nuclear reactions.

#### 6.4 Future applications of the positive ions auto-extraction

The proposed method still requires to be able to present well separated orbits at extraction to reach a clean extracted beam. In this respect it is not superior to the classical ESD extraction. But the main advantage of the proposed method is that the fragile septum of the deflector, susceptible to damage when hit by high beam currents, is now replaced by an immaterial separatrix between the internal and extracted beam. This separatrix is the  $N = -1$  line. Protons found on or close to this line will still need to be removed from the extracted beam by beam scrapers, but this beam scraping can take place downstream, where the beam density is much smaller. As these beam scrapers can be made of low activation materials, and well cooled, this extraction method allows the extraction of practically unlimited beam intensities. The suppression of the ESD is also a welcome simplification, and is likely to have a positive impact on the cyclotron reliability.

Provided that experiences confirm our numerical simulations, we think that this new extraction method is likely to replace soon the use of negative ions in cyclotrons designed for high currents. This should be the case of the 30 MeV cyclotrons used for radioisotope production, of the small cyclotrons designed for the in-hospital production of PET isotopes, and of the very large, high current cyclotrons envisioned to drive subcritical nuclear reactors.

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