# CYCLOTRON TECHNOLOGY AND BEAM DYNAMICS FOR MICROBEAM APPLICATIONS

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#### Abstract

We have been improving a beam quality of the TIARA cyclotron to form a heavy-ion microbeam with a spot size about 1  $\mu$ m. An energy spread  $\Delta E/E$  of the beam on the order of  $10^{-4}$  is required for eliminating chromatic aberrations in the focusing magnets. A flat-top acceleration system using the fifth-harmonic frequency was installed in the cyclotron to reduce the energy spread. In addition, a magnetic field stabilization system, an acceleration phase control technique and a new central region were developed to provide the microbeam stably for beam users. A cocktail beam acceleration technique was introduced to quickly change the microbeam to the other one, and a few microbeams can be used in a beam time.

## **INTRODUCTION**

Takasaki Ion accelerators for Advanced Radiation Application (TIARA) facility of the National Institutes for Quantum and Radiological Science and Technology (QST) was constructed to provide high-energy ion beams mainly for research in biotechnology and materials science. QST [1] was newly established in April 2016 by merging the National Institute of Radiological Sciences (NIRS) with a few research institutes of the Japan Atomic Energy Agency (JAEA).

An AVF cyclotron with a K-value of 110 [2] and three electrostatic accelerators are installed in TIARA, and ion beams with wide ranges of energy and ion species are available. A microbeam with a spot size about 1 µm is a powerful tool to analyze and/or irradiate a microscopic area. At TIARA, microbeam applications such as in-air Particle Induced X-ray Emission (PIXE) analysis and Proton Beam Writing (PBW) are carried out by focusing ion beams accelerated by the electrostatic accelerators [3]. On the other hand, in a vertical beam line of the cyclotron hundreds MeV heavy-ion microbeam irradiation to living cells is carried out by using micro collimators for elucidation of cellular radiation response [4]. However, spot size of the microbeam is larger than that of the electrostatic accelerator's one due to fabrication limit of the collimator and scattered ions at the edge of the collimator. In addition, targeting speed is too slow since a targeting point is adjusted by moving a mechanical sample stage. To form a microbeam with a spot size about 1 µm, a microbeam formation system using quadrupole magnets with a beam scanner was installed in the other vertical beam line of the cyclotron.

To form such a microbeam using the focusing magnet,

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an energy spread  $\Delta E/E$  of the ion beam must be reduced to the order of  $10^{-4}$  for eliminating chromatic aberrations in the focusing magnet. However, the energy spread of the cyclotron beam is typically on the order of  $10^{-3}$  while that of the electrostatic accelerator on the order of  $10^{-4}$ . A flat-top (FT) acceleration system using a fifth-harmonic frequency was developed to reduce the energy spread. In addition, cyclotron technologies such as magnetic field stabilization system and beam phase control techniques were introduced to ensure the effect of the FT acceleration. In this paper, we briefly describe the above cyclotron development, and also mention a technique to quickly change ion species of the microbeam and recent microbeam development of a 320 MeV  ${}^{12}C^{6+}$ .

# MICROBEAM FORMATION AND SINGLE-ION HIT CONTROL SYSTEM

Figure 1 shows the system of the focusing microbeam formation and single-ion hit, which means irradiating a targeted point with a high-energy ion one by one. Details of the microbeam formation system [5] are shown in Fig. 2. The system consists of quadruplet quadrupole magnets, a pair of micro slits, a pair of divergence angle defining slits, and so on. The beam shifter upstream the micro slits matches the incident beam trajectory with the microbeam line axis. Magnification factors of the focusing system for x and y directions are equally 1/5. The 90° bending magnet doesn't have a role as an energy analyser; therefore reduction of the energy spread of the incident beam is indispensable condition. The targeting point is determined by the electrostatic beam scanner. Ions pene-



Figure 1: Layout of equipment for microbeam formation and single-ion hit control system at the TIARA cyclotron facility.

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Figure 2: Beam optics, schematic view and picture of the vertical microbeam formation system. The picture was taken peering up at the target station and the Q magnets.

trating a thin target, such as living cells on a plastic film, are detected by a solid-state detector (SSD), and a fast beam kicker (P-chopper) installed in the beam injection line [6] controls a total number of ions to be irradiated as shown in Fig. 1. Fast single-ion hit of 600 samples/min is an achievable objective by using the beam scanner and the P-chopper.

To irradiate living cells, the microbeam is extracted in the atmosphere through a vacuum window made of  $Si_3N_4$ with a thickness of 200 nm. Although the vacuum window is set at the focal point of the Q-magnets the microbeam size is deteriorated because of scattering in the  $Si_3N_4$  and the atmosphere layers. For example, a 260 MeV <sup>20</sup>Ne<sup>7+</sup> is enlarged by about 2 µm at a distance of 1 mm from the vacuum window. The target sample must be placed as close to the vacuum window as possible. A microbeam irradiation experiment in vacuum is possible by exchanging the target station to the other one.

## FLAT-TOP ACCELERATION SYSTEM

To achieve an energy spread of  $10^{-4}$  an FT acceleration system using the fifth-harmonic frequency was developed for the TIARA cyclotron [7, 8] by reference to the FT system developed at the iThemba LABS for their injector AVF cyclotron with the *K*-value of 8 [9]. Figure 3 (upper) shows the picture of the TIARA cyclotron incorporated with a compact FT resonator. The FT resonator covers a wide range of resonance frequency from 55 to 110 MHz so that the FT acceleration is performed for all ion beams accelerated at the fundamental frequency ranging from 11 to 22 MHz. The fundamental and the fifth-harmonic frequency voltages are generated together on a dee electrode since there is no individual resonator for the harmonic frequency in contrast to a ring cyclotron. Figure 3 (lower) shows an example of the FT voltage waveform observed through a capacitive voltage pick up circuit. The fifthharmonic voltage, however, has significant variation



Figure 3: Picture of the TIARA cyclotron incorporated with the FT resonator (upper). Flat-top voltage waveform monitored at the dee voltage pick up (lower). The fundamental frequency (20.417 MHz) voltage was 40 kV.

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along the acceleration gap depending on the resonance frequency [8]. The voltage decreases from the tip of the dee electrode toward the outer radius. On the other hand, in the case of the fundamental frequency, negligible voltage distributions were measured. The optimum fifthharmonic voltage, therefore, was calculated at each frequency to uniform the overall energy gain of the beam bunch just before extraction from the cyclotron.

#### CONTROL OF ACCELERATION PHASE AND BEAM PHASE WIDTH

The beam bunch must be kept within the uniform region of the energy gain for successful FT acceleration. In other words, acceleration phase and beam phase width must be carefully controlled. To measure the acceleration phase in a cyclotron, there is a well-known method studied by A.A. Garren and L. Smith [10]. To easily measure and adjust the acceleration phase a new method was developed by us [11] and briefly described below.

Here we assume that no obvious voltage distribution exists along the acceleration gap and an isochronism is quite good. The internal beam current is measured at a fixed radius before extraction by a radial probe for slightly increased or decreased acceleration frequency to find



Figure 4: Expected beam current pattern in the cyclotron when the acceleration frequency is scanned. The acceleration phase is lagging in this example.



Figure 5: Beam current patterns of the 220 MeV  ${}^{12}C^{5+}$  obtained at a radius of 865 mm before extraction scanning the acceleration frequency. The beam current was normalized at  $\Delta f/f = 0$ .

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out the current pattern till the beam current decreases to the halves, where we estimate the beam bunch reaches at the energy gain of 0, as shown in Fig. 4. The acceleration phase  $\theta$  is recognized by analyzing symmetry of the beam current pattern with Eq. (1),

$$\theta = Sin^{-1} \left( 1 - 2 \frac{\Delta f_A}{\left(\frac{\Delta f_A}{f} - \frac{\Delta f_B}{f}\right)} \right).$$
(1)

Once the acceleration phase has been measured, we can optimize it by changing magnetic field of the central bump as shown in Fig. 5. The acceleration phase was estimated to be 9° advancing for the original condition; therefore the central bump was slightly decreased to optimize the acceleration phase.

In addition to the conventional method using a phase slit for restricting the beam phase width, a new technique to reduce the beam phase width using a phase bunching effect at the first revolution in a central region was developed by our group [12]. Equipment in the central region, such as an rf shielding cover of inflector electrode, a puller electrode and the phase slits were fully renewed. In particular for acceleration harmonics h = 2 the beam phase width can be reduced to about 10° or less by combination of an external beam buncher, without beam cutting by the phase slits [11]. On the other hand, the phase bunching effect has no effect for h = 1 beam of the TI-ARA cyclotron as described in the last section and the beam phase width should be restricted with the phase slits.

## MAGNETIC FIELD STABILIZATION SYSTEM

To regulate the acceleration phase at the optimum point, the magnetic field is needed to be extremely stabilized. The total amount of shift of the acceleration phase reaches tens of degrees in the case of  $\Delta B/B = 1 \times 10^{-4}$  for the cyclotron. The magnetic field of the cyclotron, however, gradually changed for long duration over 10 h on the order of  $10^{-4}$  because of heat deformation of the iron yoke caused by heat transfer from a pair of main coils. Variable temperature water-cooled copper plates were installed between the main coil and surface of the magnet yoke to insulate the heat as shown in Fig. 6. The temperature of the cooling water is adjusted between 22 to 25°C by 0.1°C as a function of the main coil current. As a result, the magnetic field instability was remarkably improved to  $\Delta B/B = 1 \times 10^{-5}$  for all ion beams [13].

Simultaneously with the stability improvement, a nuclear magnetic resonance (NMR) probe was developed to precisely measure the magnetic field in the cyclotron. An NMR probe is generally used to precisely measure a uniform magnetic field, and it was difficult to obtain sufficient signal intensity in the non-uniform cyclotron magnetic field. To improve a signal-to-noise ratio (S/N) of the NMR probe, a series of actions such as searching proper



Figure 6: Schematic view of the cyclotron magnetic field stabilization system. The system consists of the watercooled copper plates, installed between the main coil and the magnet yoke, and the additional coil driven by the PID controller. The NMR probe was placed on a central axis of a sector part of the magnet at a radius of 660 mm.



Figure 7: Improvement of the magnetic field stability by using the NMR feedback system. Black solid line was obtained by smoothing. The magnet was excited for acceleration of the 260 MeV  $^{20}$ Ne<sup>7+</sup> beam (B = 2.05 T).

installation position, vibration insulation of the signal pickup cable and addition of a noise filter circuit was carried out.

On the other hand, short duration change of the magnetic field appeared probably due to the stability of the main coil power supply of  $\Delta I/I = \pm 1 \times 10^{-5}$ . An active control system of the magnetic field using the NMR probe was developed to improve the stability more as shown in Fig. 6. The magnetic field is regulated by using an additional 5 turns coil wound along the upper main coil, a power supply and a PC-based proportionalintegral-derivative (PID) controller. Figure 7 shows an example of the change of the magnetic field with or without the feedback control system. The magnetic field was obviously stabilized by using the active control system.

## **COCKTAIL BEAM ACCELERATION**

A 260 MeV  $^{20}$ Ne<sup>7+</sup> (h = 2) microbeam with the spot size and targeting accuracy of less than 1 µm was successful formed in vacuum [5] as a result of the cyclotron development. The energy spread of the beam was reduced to  $5 \times 10^{-4}$  by the FT acceleration. Other ion beams of h =2 such as a 220 MeV  ${}^{12}C^{5+}$  and a 400 MeV  ${}^{56}Fe^{15+}$  were preliminarily developed and focused to a few µm.

Beam users of the TIARA need several kinds of microbeams providing a wide range of Linear Energy Transfer (LET) in a beam time. However, it takes about 8 h to form the microbeam including tune of an ion source, the cyclotron magnetic field, the FT system and the microbeam formation system. A cocktail beam acceleration technique was applied to the microbeam formation for quick change of the ion species [14]. This technique can change the ion species having almost the same mass-tocharge (M/O) ratio quickly by slightly adjusting the acceleration frequency or the magnetic field. The M/O resolution R is defined by Eq. (2) and the value of the cyclotron is estimated to be about 3300,

$$R = \left| \frac{(M/Q)}{\Delta(M/Q)} \right| = \left| \frac{f_{\rm RF}}{\Delta f_{\rm RF}} \right|.$$
(2)

Table 1 shows an example of the ion species with the M/Q $\approx 2.86$  that the cyclotron is able to completely separate. In the cocktail beam acceleration, magnetic rigidities of the ion species are identical; therefore, lens parameters of the beam transport line need not to be changed. Once a microbeam is formed, another microbeam of different ion species is formed in a short time. After forming the  $260 \text{ MeV}^{20}\text{Ne}^{7+}$  microbeam, the beam was changed to the <sup>40</sup>Ar<sup>14+</sup> by changing the acceleration frequency. Corrections of steering magnets in the beam transport line and a shifter magnet of the microbeam formation system were required to match the beam trajectory with the optical axis. As a result, the microbeam was changed in half an hour, and successfully provided for an experiment without deteriorating the beam spot size [15].

Table 1: Ion Species with the  $M/Q \approx 2.86$  for the Cocktail Beam Acceleration. For calculation of the M/Q, mass of the stripped electrons and the mass excess of atom were corrected.

Ion	M/Q	$\Delta(M/Q)/(M/Q)$	RF (MHz)	LET in water (keV/µm)
<sup>14</sup> N <sup>5+</sup>	2.80007	$-1.942 \times 10^{-2}$	17.8210	186.6
<sup>20</sup> Ne <sup>7+</sup>	2.85551	0	17.4750	387.2
$^{40}{\rm Ar}^{14+}$	2.85391	$-5.603 \times 10^{-4}$	17.4848	1143

## **MICROBEAM FORMATION OF CARBON BEAM** (H = 1)

The spot size of the microbeam is estimated by analyzing a secondary electron (SE) image of a copper grid with 1000 lines/in. as described in Ref. 5. The accuracy of the spot size estimation depends on the S/N of the SE image. In the case of a 320 MeV  ${}^{12}C^{6+}$  (h = 1) microbeam formation, however, the S/N adequate for the estimation could not be obtained because of poor yields of SE since the LET and the beam intensity of this fully-stripped ion was lower than the other microbeam such as the 260 MeV

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Figure 8: Formation of the 320 MeV  ${}^{12}C^{6+}$  microbeam in the air without the FT acceleration. a) Scintillation image obtained by adding up 100 pictures in which several luminescence points were observed. b) Photomicrograph of the CR-39 detector etched by a solution after  $5 \times 5$  single-ion hits.

 $^{20}\mathrm{Ne^{7+}}$  . Therefore, a new system for estimating the beam size was constructed by using an EMCCD camera with high detection sensitivity and a gadolinium pyrosilicate (Gd<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>:Ce, GPS:Ce) scintillator. This system can detect the single-ion hit position and the whole beam image is obtained by adding up individual images in which several ion hits are observed. Figure 8 a) shows the whole beam image of the 320 MeV  ${}^{12}C^{6+}$  microbeam just behind the vacuum window. The beam sizes estimated from the intensity distribution were 8.1 and 6.5  $\mu$ m (FWHM) for x and y directions, respectively. Since then, the single-ion hit irradiation on a CR-39 solid-state nuclear track detector was done in a pattern of  $5 \times 5$  points with 50 µm pitch as shown in Fig. 8 b). The CR-39 was placed in the air at a distance of 1 mm from the vacuum window. The targeting accuracy was estimated to be 7.6 µm (FWHM) by analyzing etched pit positions shown in Fig. 8 b) and there is no great difference compared with the beam spot size. This microbeam was preliminarily used to irradiate living microscopic worms.

The beam phase width of the  ${}^{12}C^{6+}$  beam was not restricted by the phase slit not to decrease the beam current at the cost of reducing the energy spread. The beam phase width was estimated to be about 20°, and was wider than the h = 2 beam such as the 260 MeV <sup>20</sup>Ne<sup>7+</sup> since no phase bunching affects for h = 1. The beam intensity produced by the ion source and transmission efficiency from the ion source to the entrance of the microbeam formation system must be increased to adequately restrict the beam phase width by the phase slit. The microbeam size will be reduced down to a few µm by the FT acceleration.

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