PRODUCTION OF MULTI-CHARGED IONS
FOR EXPERIMENTAL USE AT HIMAC

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Abstract
Since 1994, heavy-ion radiotherapy using carbon ions is successfully carried out with the Heavy Ion Medical Accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). HIMAC is dedicated to radiotherapy, but it has as a second essential task to operate as a users facility. In that scope it accelerates many ion species for basic experiments in e.g. biomedical and material science, physics and chemistry. In order to serve all HIMAC users at best, the extension of the range of ion species is an important subject in ion source development at HIMAC. Several developments on the 18GHz ECR ion source (called NIRS-HEC) are now in progress. In order to increase the beam intensity for heavier ions, additional microwave power is applied at a different frequency by a traveling wave tube amplifier. Various compounds are employed for the production of metallic ions by the metal ion volatile compound (MIVOC) technique. Results of recent developments are reported.

INTRODUCTION
Status of heavy-ion radiotherapy
Heavy-ion radiotherapy has physical and biological advantages over other types of radiation therapies. The physical advantage is a localized dose distribution just on a tumor in a human body. The biological advantages are a large relative biological effectiveness (RBE) due to the high linear energy transfer (LET), a small oxygen enhancement ratio (OER), and a small dependence on the cell cycle. Although LBL carried out pioneering trials in the 1970’s – 1980’s[1], it was not completed clinically. In order to verify the effectiveness and safety of heavy-ion radiotherapy clinically, the first medical dedicated heavy-ion accelerator in the world, named HIMAC, started operation at the National Institute of Radiological Sciences (NIRS) in 1993[2]. Over 4000 cancer patients have already been treated since 1994. These clinical results have clearly verified the advantages of heavy-ion therapy. The detailed results, i.e. a 5-year survival ratio, a local control ratio, grading of any side effect, and so on, are given in reference [3].

Based on 10 years-experience at HIMAC, a hospital-specified facility optimized for carbon ions has been designed[4]. The prototype injector, which consists of an ECR ion source (called Kei2[5]), a RFQ linac, and an IH linac[6], has been successfully developed at NIRS. Thus, in co-operation with NIRS, Gunma University has been constructing the carbon-therapy facility since April 2006. The first clinical trial is scheduled for FY2009.

Motivation of developments for ion sources
HIMAC is dedicated to radiotherapy, but - as mentioned above - it has as a second essential task to operate as a users facility. In that scope it accelerates - during evening, night and weekend- various ion species for basic experiments. Two ECR ion sources and one PIG ion source are installed in HIMAC at present. A 10 GHz ECR ion source, so called NIRS-ECR, has satisfied the medical requirements[7]. A PIG ion source[8] produces ion species from solid materials by the sputtering technique. Other ion species are supplied using the 18 GHz ECR ion source, so called NIRS-HEC[9,10]. In order to serve all HIMAC users at best, the extension of the range of ion species is an important subject in ion source development at HIMAC. The requirement for ion sources is to produce ions with a charge-to-mass ratio over 1/7 and an injection energy of 8 keV/n. The present records of beam intensities are summarized in Table 1. However, more ion species and higher intensities for heavier ions are still required. Developments for ion sources are in progress.

Table 1: Records of beam intensities without afterglow

<table>
<thead>
<tr>
<th>Ion</th>
<th>Output (μA)</th>
<th>Output (μA)</th>
<th>Output (μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂⁺</td>
<td>3000</td>
<td>P Ne⁺⁺⁺⁺</td>
<td>6000</td>
</tr>
<tr>
<td>He⁺</td>
<td>6000</td>
<td>P Mg⁺⁺⁺⁺⁺⁺</td>
<td>400</td>
</tr>
<tr>
<td>B⁺⁺⁺⁺⁺⁺</td>
<td>150</td>
<td>P Si⁺⁺⁺⁺⁺⁺</td>
<td>250</td>
</tr>
<tr>
<td>C⁺⁺⁺⁺⁺⁺</td>
<td>470</td>
<td>E Cl⁺⁺⁺⁺⁺⁺</td>
<td>400</td>
</tr>
<tr>
<td>N⁺⁺⁺⁺⁺⁺</td>
<td>1050</td>
<td>H Ar⁺⁺⁺⁺⁺⁺</td>
<td>1100</td>
</tr>
<tr>
<td>O⁺⁺⁺⁺⁺⁺</td>
<td>930</td>
<td>H Ca⁺⁺⁺⁺⁺⁺</td>
<td>250</td>
</tr>
</tbody>
</table>

Scope of the developments
In order to increase the NIRS-HEC beam intensity, the optimization of the extraction configuration was most...
effective in our case. We optimized the position the movable electrode following the simulation study[11], increased the extraction voltage[9], and optimized the radial magnetic field[10]. These results for most gaseous ion species were generally satisfying.

In addition, several techniques (tricks) are very useful to shift toward the higher charge of the charge-state distribution (CSD). That is the afterglow mode[12], the gas mixing[13], the biased electrode[14, 15, 16], and so on. The gas mixing technique is very sufficient and convenient, because it can be utilized while maintaining the present ion source structure with proven good performance, i.e. reproducibility and stability.

For the metallic ions, the effort generally is to make gas vapour from a solid material. The metal ion volatile compound (MIVOC) technique is very easy and useful way to produce some elements[17]. Below we will report on recent experimental results. Toxic (or caustic) gasses like chloride gasses are also easy to ionize, but they should of course be handled with extreme care. We developed a special gas feeding system with a gas eliminator and utilized them[18].

Unfortunately, the intensities for most heavier ion species obtained in this way are not enough for an effective extension of the range of ion species. In particular, the plasma maintained by MIVOC contains many impurities like carbon, and it disturbs effectiveness of the gas mixing. It is necessary to develop a technique to improve of CSD for heavier ions and to supply various ions.

For such ‘dirty’ gasses, the development of biased electrodes stimulating an ‘anti-wall coating’ effect is planned[7]. The Bio-Nano ECRIS at Toyo Univ.[19] is assigned as a test bench under the cooperation. Two-frequency heating appears to be also a promising technique to handle such a dirty plasma. We will report recent experimental results in this paper.

It is, of course, important to develop new techniques for the production of pure plasmas in a more general way for every solid material. An electron bombardment evaporator and / or an induction heating oven for high temperature are being planned for that purpose.

**EXPERIMENTAL RESULTS OF TWO-FREQUENCY HEATING**

**Summary of former experiences**

Many reports pointed at the improvement of highly charged ion production by feeding multiple microwaves with different frequencies[20]. In an early stage of the development of two-frequency heating using two klystron amplifiers (KLY), we could confirm that the two different frequency microwaves were absorbed at different ECR zone by observing the shapes of visible radiations[21]. Although the beam intensity was improved, the tuning of operation parameters was difficult and not so reliable. We thought the reason was due to interference of one microwave with the other microwave.

The KLY had over 1 kW power, but its frequency was fixed. In order to investigate the frequency dependence precisely, we added an additional travelling wave tube amplifier (TWT) with a wide frequency range between 10 and 18 GHz. A hyperfine structure appeared in the frequency dependence of the additional microwave[22]. Three major parameters for operation of ECRIS, i.e., the vacuum pressure, the magnetic confinement, and the power of the microwave, are correlated to each other. Although the frequency of microwave changed the plasma condition, the optimizing of other two parameters usually recovered from the bad condition. On the other hand, since the magnetic confinement can not be optimized in the case of the permanent magnet ECRIS, it was very important to tune the frequency. In the case of two-frequency heating, it is also important to adjust one frequency to the fixed condition given by the other frequency. Our TWT had only a maximum power of 250 W at the former experiments. As a result, it was not enough to improve the performance. Therefore, the conclusion of our initial experiences is that a TWT with fine tuning of frequency and enough power is required.

**Experimental setup**

The experiments are being performed with NIRS-HEC. NIRS-HEC was designed to reach a high extraction voltage and a high magnetic field with normal conducting magnets. The maximum extraction voltage between the plasma slit and the extraction electrode is 60 kV. The maximum mirror fields at the injection and at the extraction side are 1.3 and 1.2 T, respectively. The microwave power is usually supplied by an 18.0 GHz KLY with a maximum power of 1.5kW.

![Experimental setup](image)

Figure 1: Circuit diagram of the additional microwave system.

![Experimental setup](image)

Figure 2: Layout of waveguides.

In order to investigate the effects of two-frequency heating, an additional microwave-injection system was added to NIRS-HEC which has a frequency range of 17.75 to 18.25GHz. The system consists of a frequency synthesizer, two 500W TWT and a waveguide system. The waveguide system has a power combiner and a feedback monitor to stabilize the forward power in the full frequency range. The maximum input power is over 600W at the entrance into the ion source’s chamber.
Figure 1 shows a circuit diagram of the additional microwave system. The microwaves from KLY and from TWT are injected into the plasma chamber by two rectangular waveguides at the RF shielding endplate shown in Figure 2.

Experimental result

Experimental data were obtained with $^{84}$Kr gas (isotopic enrichment 80%). Initially, the frequency and microwave power from the TWT were set at 18.0 GHz and 600 W, respectively. The other parameters, i.e., amount of gas, magnetic field, extraction voltage, and so on, were optimized. Then, the frequency dependence of the output current of Kr$^{15+}$ was measured as shown in Figure 3 (solid line). The phenomena of frequency dependence have been studied by many groups, and they were very complicated (for a recent example see ref.[23]). In our case the effect case was related to a shift in the CSD. Mass spectra which were measured at three different frequencies clearly show that the CSDs varied with frequency in Figure 4.

![Figure 3: Frequency dependence of Kr$^{15+}$ output current. The solid line and broken line are obtained by TWT only and TWT+KLY, respectively. The asterisk mark is obtained by KLY only.](image)

When TWT was stopped and only KLY supplied 600 W, the beam intensity was decreased as shown in Figure 3 (asterisk). When the microwave power from KLY was increasing, the output current was also increasing. However, the beam instability was appeared over 700 W. Figure 5 shows measured time structures of beams with different microwave powers, 600, 800 and 900 W. The plasma collapsed every several milliseconds and recovered in about 1 ms. When TWT turned on in this situation, the beam stability was improved and the large output current was obtained as shown in Figure 5. The frequency dependence with TWT+KLY was also appeared as shown in Figure 3 (broken line), but its fluctuation became more calmly. It is noted that each microwave power does not equal exactly due to loss in the waveguides.

![Figure 5: Time structures of beams.](image)

As a conclusion, the double frequency heating improved the beam intensity under the conditions of enough power and precise frequency tuning for the additional microwave. It seems it is mainly due to prevent the plasma instability in our cases. The maximum record of output current for $^{84}$Kr$^{15+}$ was 205 $\mu$A (80% enrichment). This record was about 30 % larger than the
record with the single frequency heating, and its beam stability was more stable. The optimized microwave power is not saturated. Applying a more powerful TWT would be promising. For higher charge state ions, it is more effective as shown in Figure 6.

**EXPERIMENTAL RESULTS OF MIVOC**

The MIVOC - method is used for the production of various ion species in Table 2. In order to obtain the desired evaporation and consumption rates in each case, the temperatures of the MIVOC oven and of the gas-feeding line are controlled independently. A thermal-control system has been developed for utilizing metal ion volatile compounds with various vapour pressures.

For low vapour pressure like (C\textsubscript{2}H\textsubscript{2})\textsubscript{2}Mg, a thermostat is heating a container with the compound up to a suitable temperature. The gas feeding tube must be kept higher temperature than the container to prevent condensation or adsorption to the tube’s wall. On the other hand, for high vapour pressure like (CH\textsubscript{3}C\textsubscript{2}H\textsubscript{2})\textsubscript{4}Co, the compound must be cooled lower than the room temperature. A cryostat with Peltier elements is utilized here.

<table>
<thead>
<tr>
<th>Ion</th>
<th>Compound</th>
<th>Output (e\textmu A)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{24}\text{Mg}^{+})</td>
<td>(C\textsubscript{2}H\textsubscript{2})\textsubscript{2}Mg</td>
<td>150</td>
<td>Necessary to heat</td>
</tr>
<tr>
<td>(^{28}\text{Si}^{+})</td>
<td>Si(CH\textsubscript{3})\textsubscript{4}</td>
<td>250</td>
<td></td>
</tr>
<tr>
<td>(^{56}\text{Fe}^{9+})</td>
<td>(C\textsubscript{2}H\textsubscript{2})\textsubscript{2}Fe</td>
<td>400</td>
<td>Better with heating</td>
</tr>
<tr>
<td>(^{59}\text{Co}^{9+})</td>
<td>(CH\textsubscript{3}C\textsubscript{2}H\textsubscript{2})\textsubscript{4}Co</td>
<td>170</td>
<td>Necessary to cool</td>
</tr>
<tr>
<td>(^{74}\text{Ge}^{12+})</td>
<td>GeH(\text{CH}_{3})\textsubscript{3}</td>
<td>50</td>
<td></td>
</tr>
</tbody>
</table>

**FUTURE PLANS FOR PURE METALLIC IONS**

**Induction heating oven**

MIVOC is a useful technique for several metallic ions. However, it is necessary to make efforts for developments of each new ion species, and there is the well known disadvantage that inherently many contaminations are included in the production. Carbon ions from many compounds (See Table 2.), due to their deposition on the wall or an insulator, especially disturb application of other useful tricks like wall coating or biasing electrodes. Also, it is difficult to optimize the amount of support gas for gas mixing. In order to prevent such disadvantages and to produce a “more pure metallic gas” from various materials, an induction heating oven is under considered.

The Osaka University group has developed such an oven system[24] that could be tested at Kei2[25]. We have a plan to use this type of oven to NIRS-HEC.

**Electron bombarder**

In order to produce more pure metallic gas from especially high melting point materials, a gas supply or feed method by electron bombardment is under development. A metallic target rod or crucible at a high positive potential is heated by the electron bombardment technique. The temperature of materials easily reaches more than 2000 °C. Although we obtained an output current of 25 e\textmu A for Fe\textsuperscript{9+}[26], the evaporation rate and the lifetime are still not stable and have to be controlled better.

**REFERENCES**

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[19] T. Uchida et al., in these proceedings.