FORMATION OF HIGH-INTENSIVE RADIOACTIVE CARBON ION BEAMS IN THE ELECTRON STRING ION SOURCE

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Abstract

Accelerated ^{12}C ion beams are effectively used for cancer treatment at various medical centers, in particular to treat patients with radio resistant tumors. On the other hand, positron emission tomography is the most effective way of tumor diagnostics. The intensive ^{11}C ion beam could allow both these advantages to be combined. It could be used both for cancer treatment and for on-line positron emission tomography. Formation of a primary radioactive $^{11}C^{6+}$ ion beam with the intensity of 10^{10}-10^{11} pps from the ion source may allow cancer treatment and on-line dose verification.

^{11}C isotope is produced in the nuclear reaction ^{14}N $(p,\alpha)^{11}C$ using the gas target chamber irradiated by a proton beam. If the nitrogen target chamber contains about 5% of hydrogen, approximately 10^{14} methane molecules $^{11}CH_4$ can be produced each 20 minutes. The separated radioactive methane can be loaded into an ion source.

The methodology and technique of formation of high-intensity radioactive carbon beams were tested in the JINR electron string ion source (ESIS) Krion-2 using usual non radioactive methane. The measured conversion efficiency of methane molecules to carbon ions appeared to be rather high, 15 % for C^{6+} ions and 25% for C^{4+} ions. The developed technique of pulsed methane loading and the experimentally obtained conversion efficiency permit obtaining primary radioactive $^{11}C^{6+}$ beams at the intensity of 10^{10} -10^{11} pps and performing cancer treatment and on-line dose verification.

FORMATION OF HIGH-INTENSITY RADIOACTIVE CARBON ION BEAMS

Accelerated $^{12}C^{6+}$ beams at the ion intensity of 10^9 particles per second (pps) are effectively used for cancer treatment at various medical centers, in particular to treat patients with radio resistant tumor targets [1]. On the other hand, positron emission tomography (PET) is the most effective way of tumor diagnostics.

Accelerated ion beams of the positron-emitting ^{11}C isotope (half-lifetime is about 20 min) were first used at NIRS-HIMAC for cancer therapy applications [2]. The use of the ^{11}C ion beam could allow both these advantages to be combined because this beam could be simultaneously used both for cancer treatment and for on-line positron emission tomography. Verification of the radiation dose in the tumor target will be carried out simultaneously with cancer treatment.

In order to produce ^{11}C beam for cancer therapy, the Projectile Fragmentation Method (PFM) was used at HIMAC [2]. In this scheme the ^{7}Be target was irradiated by the accelerated primary ^{12}C beam and the maximum ^{11}C production rate was about 1% and the purity was near 93%.

To increase the intensity of radioactive carbon ion beams by two orders of magnitude, the ISOLDE scheme was proposed [3]. An advantage of primary radioactive ^{11}C ion beams is the higher space resolution at PET tomography compare to secondary radioactive beams produced in PFM.

In the ISOLDE scheme ^{11}C isotope is produced through the nuclear reaction ^{14}N $(p,\alpha)^{11}C$ in the target chamber filled with N_2 gas at the initial pressure of about 20 bars. The proton beam from the 18 MeV cyclotron allows getting the activity of 1.5 Ci (10^{14} atoms of ^{11}C) for 20 min of irradiation with the proton beam current of 20 μA. The nitrogen gas target also contains 5% of H_2 instead of 2% of O_2 gas [3] to produce $^{11}CH_4$ molecules which are more applicable for our scheme. A Porapac cryogenic trap is used for separation of frozen radioactive methane $^{11}CH_4$ from N_2 gas pumped away from the trap with a flow rate of about 1.5 l/min. After stopping the flow of the target gas, the cryogenic system raises the temperature and radioactive methane is passed into a tank. In these steps, the ^{11}C collection efficiency is expected to be about 95%. The total separation time is about 3 min. After the separation about $8.5 \cdot 10^{13}$ $^{11}CH_4$ methane molecules still remain, and they could be loaded into an ion source.

The standard transmission injection-extraction efficiency of the beam at the HIMAC synchrotron is around 10%. Since the HIMAC operation is based on a very low duty factor with a range of 10^3, it is necessary to provide pulsed gas injection into the working space of the ion source. The Electron String Ion Source [5, 6] is one of the promising ion sources for generation of the positron-emitting $^{11}C^{4+}$ ion beam at the intensity of 10^{10} pps. The $^{11}C^{4+}$ ions formed in the ion source could be injected in the linac at the repetition frequency of 1 Hz, where after acceleration up 6 MeV/n, they will be transformed into $^{11}C^{6+}$ ions on a target and then will be injected in HIMAC. Finally, the ^{11}C ion beam at intensity about 10^9 pps will be extracted from HIMAC on a tumor target.

Realization of such parameters could provide the possibilities of the synchrotron-based cancer treatment and on-line dose verification both on the existing carbon accelerators and on the projected ones. This work is a step towards this goal.

DECAY AND INJECTION OF ^{11}C ISOTOPES

The number of the remaining $^{11}CH_4$ molecules (Figure 1) decreases with time because of their

radioactive decay and also because of their pulsed injection into the electron string ion trap

$$N(t) = N_0 \cdot \exp(-\lambda t) - (\dot{n}/\lambda\xi) \cdot [1 - \exp\{-\lambda t\}],$$

where N_0=8.5·10^{13} is the initial number of $^{11}CH_4$ molecules loaded in the ion source, $\dot{n}$ =10^{10} pps is the ion injection rate, λ=1/1200 s^{-1} is the decay constant, ξ=0.25 is the conversion efficiency of the methane molecules to ions.

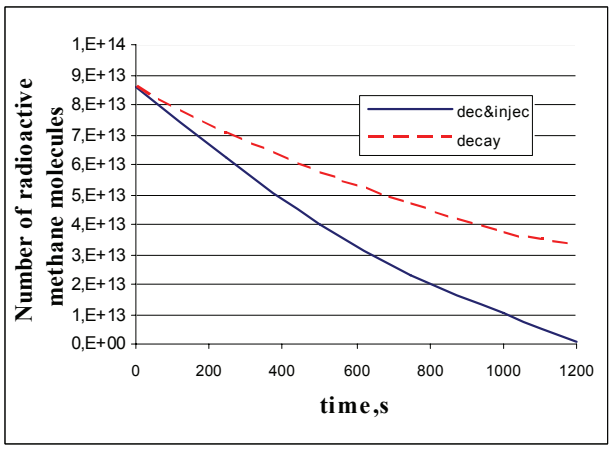

Figure 1: Dependences of the number of remaining $^{11}CH_4$ molecules on time (upper curve: radioactive decay alone; lower curve: decay and pulsed injection).

The remaining number of $^{11}CH_4$ molecules after 20 min injection is still large, N=10^{12}. The injection efficiency corresponding to the ratio of the number of $^{11}C^{4+}$ ions injected during the cycle to the number of methane molecules loaded in each cycle is $\eta = \dot{n}/(N_0 \cdot \lambda) = 0.12$ at the parameters given in Fig. 1.

ELECTRON STRING ION SOURCE

The Electron Beam Ion Source (EBIS) [7] usually operates in the "direct beam" mode in order to produce highly charged ions. The dense electron beam of about 1 m long and of about 1 mm in diameter, which is confined in such a source by a strong longitudinal magnetic field of a super conducting solenoid, is used both for confinement of positive ions in the beam electron space charge and for step-by-step electron impact ionization of the ions. The highly charged ions can be extracted from the source and used as required. High power of the electron beams and especially their high power density in the electron collector region are disadvantages of EBIS sources.

The reflex mode of the EBIS operation (ESIS mode) [5, 6, 8] is realized by using the specially designed electron gun and the electron reflector that allows a multiple use of beam electrons. The electrons do not reach the electron collector after one pass through the drift space of the source; instead, they are reflected backwards to the emitter side and then are reflected again in the vicinity of the emitter and so on. Finally, the reflected electrons can perform, depending on experimental conditions, up to 10^3 oscillations between the electron emitter and the collector. When due to the reflections the electron density reaches a definite value, a phase transition can occur to the so-called electron string state. The string electrons are used for ion production in ESIS (Table 1) similar to the beam electrons in EBIS. And in the ESIS mode it is possible to reduce the power consumption of the ESIS high-voltage system by 1 - 3 orders of magnitude and to obtain a similar effective electron density, which allows improving the source reliability and at the same time increasing the ion output. As a result, the power of the high-voltage system in ESIS corresponds to several tens Watts.

The electron string phenomenon was first discovered at JINR LHE about 10 years ago and after sufficient researches the Krion-2 ESIS was constructed. Now it is successfully used as the ion source producing highly charged ions beams for the JINR relativistic super-conducting synchrotron Nuclotron [9]. This source provides a rather high pulse intensity of really highly charged ion beams, for example, Ar^{16+} - 200 μA, Fe^{24+} - 150 μA in 8 μs pulses.

Figure 2: Electron String Ion Source Krion-2.

Table 1: Parameters of Electron String Ion Sources

Ion source	Krion-2 C^{4+}	Krion-5T C^{4+}	TESIS C^{4+}
Electron energy, keV	3-5	5-7	5-7
Number of electrons	6·10^{10}	3·10^{11}	3·10^{12}
Magnetic field, T	3	5	5
Ion current, mA	0.2	1	10
Pulse duration, μs	8	8	8
Number ions extracted per pulse	2.5·10^9	10^{10}	10^{11}
Injection frequency, Hz	100	100	100
Average current, μA	0.15	0.8	8

The charge capacitance of the Krion-2 ion trap is $6 \cdot 10^{10}$ elementary charges. The trap can be easily half-filled with low charge state (similar to C^{2+}) ions during 1-2 ms pulse injection, and then during about 5 ms confinement in the electron string the ions will increase their charge state by a factor 2, filling the trap and compensating the negative electron space charge. As was shown experimentally, adjusting the electron energy, injection time, and time of ion confinement, one can get up to 50 % of C^{4+} in the total ion beam pulse extracted from the source. It is pertinent to mention that hydrogen ions produced from methane molecules leave the ion trap taking energy from carbon ions which the last ones take from string electrons. This process was also studied earlier and was given the name ion-ion cooling. Increasing the confinement time to about 90 ms, one can get mostly the C^{6+} ion beam, but because of some losses during the longer confinement the number of particles will be slightly smaller than for C^{4+} beam.

So, the existing ion source Krion-2 could produce around $2.5.10^9$ C^{4+} particles per pulse. Another advantage of the source is the quite small emittance of extracted beam (5-10 π·mm·mrad).

MODELING OF FORMATION OF RADIOACTIVE ION BEAM IN ESIS

We used ordinary methane $^{12}CH_4$ for this research since the problem under consideration does not depend on the type of the carbon isotopes incorporated into the methane molecule. According to the HIMAC cancer therapy requirements, the ion source should produce C^{4+} ion beams with the intensity of 10^{10} pps and pulse width of 1 ms. Note that a limited number of methane molecules ($\sim 10^{14}$) is available during each 20 minutes. The problems we faced and solved are the following: 1) pulsed injection (in ms range) of some optimal number of methane molecules into the working space of the ESIS, keeping all the rest molecules for the next ionization cycles; 2) studies of carbon loss, caused by production of neutral radicals (CH_3, CH_2, etc) by the electron impact and their subsequent escape; 3) minimization of carbon ion loss during ionization.

CRYOGENIC PULSED GAS INJECTION TECHNOLOGY

The scheme of cryogenic pulsed gas injection [4] is given in Fig. 3: 1 is the injection section drift tube structure at temperature 78 K, 2 are the main ion trap drift tube sections at temperature 4.2 K, 3 are the ion potential barriers during ion injection, 4 and 5 are the ion potential barriers during ion confinement, 6 is the methane freezing-evaporation cell, 7 is the cell road covered by aluminized mylar, 8 and 9 are the junctions of the interior and exterior aluminum layers of mylar, 10 is the copper wire connecting the rod with the terminal at 4.2 K, 11 is the small vessel volume, 12 is the big vessel volume, 13 is the vacuum meter, 14 is the vacuum gauge, V1-V4 are the valves for methane isolation.

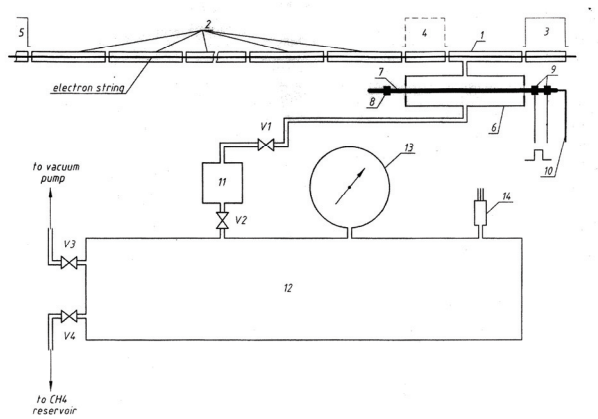

Figure 3: Scheme of Krion 2 cryogenic pulsed injection.

A special cryogenic cell was elaborated and successfully tested [4]. The cell consists of an isolated chamber which is situated in the vicinity of the working space of the ESIS, 1 cm away from the axis. The isolated chamber is arranged at the 78 K cold terminal. The key element of the cell is a copper rod 2.5 mm in diameter and 90 mm in length located in the middle of the chamber. The rod is arranged along the axis of the cell and fixed by its ends to the opposite walls of the chamber. One rod end is connected to the 4.2 K cold terminal, being thermo insulated on the chamber walls. As a result, every portion of the loaded methane molecules is completely frozen at the rod surface, which has temperature 4.2 K. The rod has sandwich-type insulation layers in its outer part and an aluminium conducting layer on its surface, which provides pulsed heating of the surface from 4.2 K to 40 ÷ 45 K during a few ms due to pulsed electrical current through the aluminium layer. As a result, all methane molecules leave the rod surface. The resulting methane vapour pressure at this temperature provides penetration of about 10^{10} molecules into the working drift tube of the ESIS. When the electrical current passed through the aluminium layer is switched off, the temperature at the rod surface decreases to 4.2 K during few ms as well and all the rest of the methane molecules are frozen again at the rod surface. Thus, the elaborated cryogenic cell provides pulsed injection of methane molecules with the pulse duration in the ms range, sufficiently reducing the total working gas (methane) consumption. Moreover, the cell works with relatively small pulse power consumption (about 40 W).

CONVERSION EFFICIENCY

The electron strings used in the Krion-2 were formed by injected electrons of energy 4.4 keV and reflection voltages about 6 kV. First, we experimentally studied the initial part of the CH_4 to C^{4+} (C^{6+}) transformation. Figure 4 shows the ion charge state spectrum when the methane injection was 30 μs long and the produced ions were extracted from the ion source without any additional confinement.

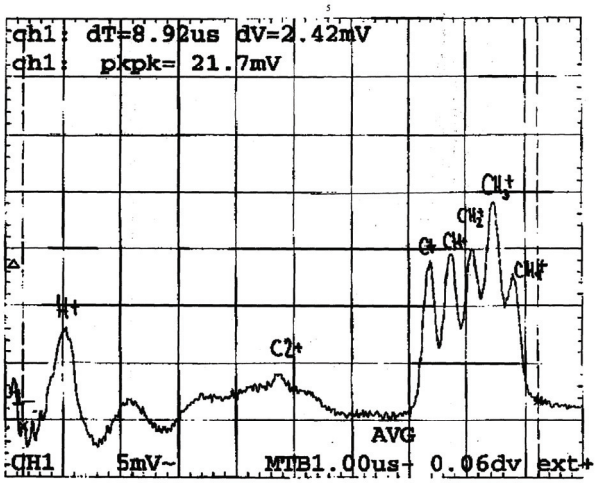

Figure 4: Ion spectra after 30 μs methane injection into the Krion-2.

One can see that in addition to H^+ it predominantly comprises singly charged methane molecule ions, hydrocarbon radical ions, and atomic carbon ions CH_4^+, CH_3^+, CH_2^+, CH^+, and C^+. In fact, neutral hydrocarbon radicals can also be produced during the initial part of the transformation. Some part of the neutrals may leave the source electron string without ionization. Therefore, the total transformation efficiency of the methane molecule to carbon ion can be decreased. It was difficult to estimate a priory the influence of the production of neutrals on the total conversion efficiency; nevertheless, it was done experimentally.

Testing various times of methane injection and ion confinement we found out that the electron string carbon ions reach the charge state C^{4+} approximately in 8 ms and the charge state C^{6+} in 90 ms. The ion charge state spectrum after 6 ms confinement is presented in Fig. 5. The ion charge state spectrum after 90 ms confinement is presented in Figure 6.

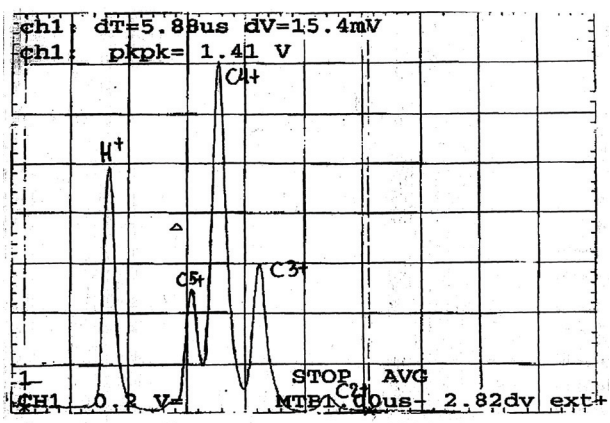

Figure 5: Ion spectra after 6 ms confinement in the electron string of the Krion-2.

Then, injecting methane molecules during 1 ms, we found out that after their confinement in the Krion-2 electron string during 5.8-6.2 ms the percentage of C^{4+} in the carbon ion charge state distribution is the highest and amounts to 53%. Injecting methane molecules during 5 ms and confining them in the electron string during 90 ms, we obtained 67% of C^{6+} in the carbon ion charge state distribution in the extracted ion beam. We consider this confinement time and percentage to be the optimum ones in production of C^{6+} ion beam. A further increase in the confinement time could increase the percentage, but the total conversion efficiency would decrease because of additional ion losses during the confinement.

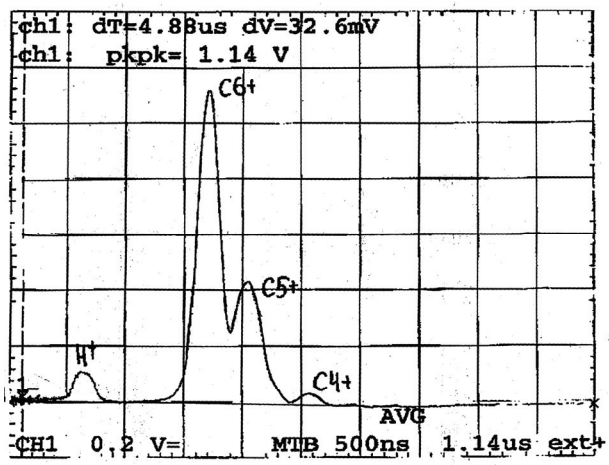

Figure 6: Ion spectra after 90 ms confinement in the electron string of the Krion-2.

As a rule, we introduced about $4 \cdot 10^{14}$ methane molecules into the cell. Then we reached the necessary heating pulse power (36 W) at 1 Hz repetition rate of heating pulses. The maximum repetition rate of accumulation – ionization was 100 Hz. In this regime we usually accumulated 26-30 nC of positive ion charge per second.

The introduced portion of methane evaporated in 2 hours, which means that in $7.2 \cdot 10^3$ seconds we produced $(1.87 \div 2.16) \cdot 10^5$ nC at the repetition rate 100 Hz. Now it is easy to calculate the conversion efficiency of methane molecules to carbon ions considering that 67% of the ion charge is the C^{6+} and 55% of carbon ions are C^{4+} ions. The number of the produced C^{4+} ions was $1.02 \div 1.18 \times 10^{14}$. This gives us the conversion efficiency $\xi = 26 \div 29\%$. To measure the conversion efficiency of methane to C^{6+} ions, we had to use 10 Hz repetition rate of 60 ms accumulation and 40 ms ionization process. In this case the conversion efficiency is evaluated to be $\xi = 12\text{-}15$ %.

We would like to summarize the conversion efficiency results:

- The cryogenic technology of accumulation and pulsed injection of methane into the electron string has been elaborated and proven experimentally.
- It has been shown experimentally that neutral CH radicals cannot dramatically decrease the conversion efficiency of methane to carbon ion beams in the ESIS.

- The measured conversion efficiencies of methane molecules to carbon ions beams is rather high, 26 – 29% for C^{4+} and 12-15 % for C^{6+} ion beams.

PERSPECTIVE OF RADIOACTIVE CARBON BEAM FORMATION

An increase up to 10^{10} of the total number of the produced C^{4+} ions per pulse (Table 1) is expected to be possible with the new Krion-5T ESIS source, which is under construction at JINR now at magnetic field 5T. It was found experimentally [5, 6] that the maximum number of electrons accumulated in a string was proportional to the confined magnetic field B to the third power $Qe=aB^3$. (Fig. 7). A grows of the magnetic field from 3 T in Krion-2 to 5 T in Krion-5T could permit about 4.5 times increase in the number of stored electrons and their density. The increase in the electron density at 5T reduces the ion confinement time, which determines the injection repetition frequency.

A further increase in the intensity of the radioactive carbon ion beams is connected with the construction of the Tubular Electron String Ion Source (TESIS) [10-11] with a capacitance of the electron string and stored ions 50 times larger of Krion-2 (Table 1). The main point is that the tubular geometry of the drift tube structure allows one to avoid virtual cathode formation for the corresponding number of accumulated electrons. The gain of the highly charged ion input in TESIS compared with ESIS is characterized by the ratio of the tubular beam diameter d to the radial beam thickness a: $N_{TESIS}/N_{ESIS}=2\cdot d/a \approx 50$. It is expected that this new TESIS will meet all rigid conceptual and technological requirements and should provide an ion output approaching 10 mA of C^{4+} ions in the pulse mode or 10^{11} ions per pulse and about 10 μA of C^{4+} ions in the average current mode.

The high conversion efficiency and the carbon ion beam intensities obtained at ESIS make it uniquely favourable for cancer treatment with ^{11}C ion beams.

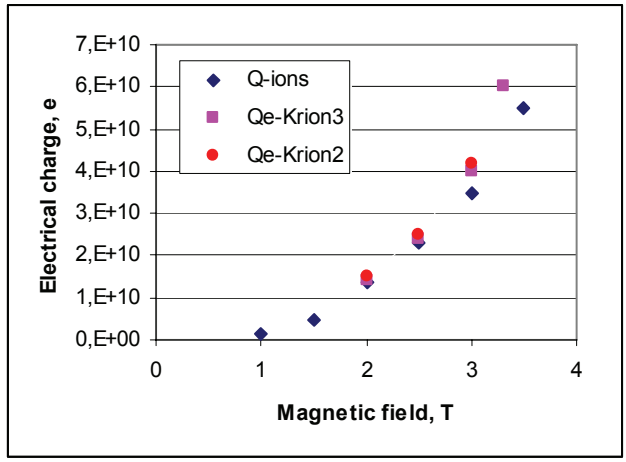

Figure 7: Dependence of the electron and ion space charge capacitances on the confined magnetic field.

ACKNOWLEDGMENTS

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