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Plasma isotope separation based on ion cyclotron resonance

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Abstract. Experiments that have been conducted in the USA, France, and Russia to investigate isotopically selective ion cyclotron resonance (ICR) as a tool for plasma isotope separation are analyzed. Because this method runs into difficulties at low values of the relative isotope mass difference $\Delta M/M$, for some elements (for gadolinium, as an example) isotope separation still remains a problem. There are ways to solve it, however, as experimental results and theoretical calculations suggest.

1. Introduction

More than thirty years have passed since the publication of the fundamental experimental work by Dawson et al. [1] on the method for plasma isotope separation based on ion-cyclotron resonance (ICR) in plasma — the ICR method. We note the preceding theoretical work by Askarian, Namiot, and Rukhadze [2], who suggested a pulsed scheme for separating ions by masses in a plasma trap and also formulated the principal conditions for their selective cyclotron heating. The start of experimental work in this field was furthered by the work done by Schmitt [3], who studied the dispersion of a kind of plasma waves (Bernstein waves) in potassium plasma and found the attenuation of these waves near the cyclotron frequencies of both the $^{39}K^+$ ion of the primary potassium isotope and the $^{41}K^+$ isotope whose concentration is lower by an order of magnitude. It was, in essence, the first observation of isotopically selective cyclotron heating in plasma that aroused practical interest. Earlier, in investigations on controlled fusion (CF), the selective cyclotron heating of only hydrogen and helium isotopic ions was performed.

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In this paper, we estimate presently known results from studying the ICR method for isotope separation. The ICR method (the method of isotopically selective ion cyclotron resonance in plasma) has not been tested in industrial conditions yet. However, experience in working with intensive separation has been acquired. The experience was gained in the USA at the PPM (Production Prototype Module) facility that was built in the early 1980s in the framework of the Advanced Isotope Separation Program financed by the US Department of Energy (DOE) and aimed at the development of a new method for uranium isotope separation in place of the gaseous diffusion separation employed in industry in the USA. The installation was built and operated for a long time by TRW (Thompson–Ramo–Wooldridge Inc.) in Redondo Beach, California. A magnetic field at the facility was produced by a large superconductive solenoid ($B \leqslant 1.8$ T).

ICR separation known in the USA as 'plasma separation process' (PSP) was not accepted as a new uranium isotope separation method because in 1982 the DOE preferred the laser method AVLIS (Atomic Vapor Laser Isotope Separation). The reasons for this choice were not made clear. We may assume that technologically this laser method had been better developed by that time than PSP. It is noteworthy that the AVLIS technology for separating uranium isotopes did not eventually become industry technology in the USA; in the late 1990s, work in this field was terminated.

Demonstration of isotope separation for stable elements [4] at PPM became a kind of answer to the negative DOE resolution. The elements included nickel, indium, and lead. The main goal was to show the possibilities of the method. Nevertheless, palladium enriched at this installation by ^{102}Pd has been applied in medicine and was used by Theragenics Corporation (TGX) [5].

PPM operation was accompanied by extensive scientific research. Report annotations of the IEEE International Conference on Plasma Science in 1983 in San Diego (including the above-mentioned report [4]) illustrate the wide range of studies. Unfortunately, no detailed publications succeeded. Several technical reports concerning this theme followed in the ensuing years. The lack of publications was compensated by a patent application by TRW [6].

Work at TRW lasted into the late 1980s. Then, the installation moved to Oak Ridge. It was supposed to apply PPM for a preliminary enrichment of the raw material used in electromagnetic separation installations operated at that place. An increase in the initial concentration of the separated isotope in a raw material by a factor of dozens might enhance the productivity of the installations by the same factor and solve, in particular, the problem of obtaining rare isotopes in the required quantity. However, these plans have not been realized. The PPM installation was not put into operation for more than ten years.

In the period of PPM conservation, experimental and theoretical works were carried out in France and Russia. The French facility ERIC (d'Experience de Resonance Ionique Cyclotron) (Louvet et al.) was located at the Saclay Research Center [7]. The facility had a superconducting solenoid with $B \leqslant 3$ T. Isotope separation of calcium, barium, gadolinium, and other elements was investigated.

In Russia, an ICR facility for separating lithium isotopes has been created at the Kurchatov Institute of Atomic Energy (Karchevskii et al. [8]). The unofficial name of the installation, Siren' (Lilac), originates from the color of lithium plasma.

An ICR facility at UCLA (University of California, Los Angeles) was built in the USA [9]. In contrast to PSP, the separation process was termed PEP (Plasma Enrichment Process). Work on rubidium isotope separation at this facility was supported by the DOE [10].

Already in the early 1990s, at a scientific conference in France the problem of obtaining gadolinium enriched by ^{157}Gd isotope via the ICR method was posed [7]. It is known that natural gadolinium is employed as a burnable additive to nuclear fuel for power reactors. Substituting it for gadolinium enriched by the mentioned isotope eliminates certain technological difficulties and may yield an economic profit [11]. There is an opinion that the ICR method for isotope separation is capable of producing isotopically enriched gadolinium in the quantity of several dozen kilograms yearly (a centrifugal method cannot be applied for enriching gadolinium because it has no volatile chemical compounds). In the framework of programs at the International Scientific-Technical Center (ISTC) in 1999 – 2000, technical project No. 0830 was fulfilled for the ICR installation MCIRI (Method of Cyclotron Ion Resonance of Isotopes) [12]. A I Karchevskii and V E Keilin (Russian Research Centre 'Kurchatov Institute') were scientific supervisors of the project. The feasibility study was performed at the All-Russian Design and Research Institute for Energy Technology (VNIPIET in *Russ. abbr.*), Saint Petersburg. By the way, the question concerning application of the ICR method for uranium isotope enrichment did not arise because the information available by that time had proved the advantage of the centrifugal method in this case.

In 2000, the DOE leased PPM to TGX Corporation, which is a producer of the medical anticancer agent TheraSeed. This drug includes the radioactive isotope ^{103}Pd which can be obtained by subjecting either monoisotopic rhodium ^{103}Rh(p,n) to proton irradiation or palladium enriched with the ^{102}Pd isotope to neutron irradiation. It was reported that obtaining ^{102}Pd in the quantities sufficient for doubling TheraSeed production was one of the aims of leasing PPM. It seems that successful experiments on separating palladium isotopes at this facility in the 1980s encouraged the leaseholder. In addition, the manufacture of various isotope products was planned [5]. The leaseholder has built a separate building for PPM at Oak Ridge.

Palladium enrichment started at TGX only in 2004. Heretofore, the DOE project (NERI Project No. 99-0074) Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors has been carried out at the leased facility. The project comprised three phases: the search for the most appropriate elements for use as burnable additives [13], then their isotope enrichment at PPM, and finally the search for an efficient variant for introducing additives into a fuel element (FE) [14]. In accordance with the findings [13], gadolinium, dysprosium, and erbium (isotopes ^{157}Gd, ^{164}Dy, ^{167}Er, respectively) were chosen for isotope enrichment.

This is the chronology of the experimental work on ICR isotope separation. These experiments will be discussed in Sections 2 – 5 along with several theoretical studies.

2. The separation process

In solving the CF problem, a great number of experimental and theoretical works are devoted to the ion cyclotron heating of hydrogen plasma, establishing a firm physical basis for cyclotron heating. However, these works did not stimulate the creation of a new method for isotope separation. The development of a new isotope separation method was retarded by the bad quality of ICR spectra of hydrogen and helium isotopic ions, as well as H^+ and H_2^+ ions. Attention should have been paid to the fact that cyclotron heating was usually employed at facilities with a nonuniform magnetic field. However, the fact is that isotopically selective cyclotron heating of plasma with heavier ions was only carried out in the middle of the 1970s [1]. By that time, the ion cyclotron resonance spectrometry had been already developed, which completed the electromagnetic mass spectrometry.

The acquired experience on separating isotopes by the ICR method confirms simple formulae concerning the isotopically selective heating of ions in plasma [1, 2]. We consider these formulae prior to describing the separation process itself.

Cyclotron heating of ions in a plasma is usually performed by a small-amplitude alternating external magnetic field $\tilde{B} \ll B$, whose oscillation frequency is chosen close to the Larmor ion frequency: $\omega_{\rm ci} = eB/M_{\rm i}$. The alternating magnetic field induces in the plasma an electric field which accelerates the ions. Since the field interacts with a moving ion, the resonance frequency is shifted relative to the Larmor frequency due to the Doppler effect: $\omega = \omega_{\rm ci} + k_z v_{{\rm i}z}$ [15]. In what follows, we suggest that the basic magnetic field is directed along the z-axis, i.e., $v_{{\rm i}z}$ is the velocity of ion motion along the magnetic field, the longitudinal velocity, and k_z is the longitudinal component of the wave vector. Only for the longitudinal temperature and energy do we use the traditional subscript $||$: $T_{{\rm i}||}$, $W_{{\rm i}||}$. Note that the subscript $\perp$ in this paper refers to the vector components in the direction normal to the magnetic field — for example, a transverse velocity, a transverse electric field, and the corresponding quantities derived from them (in particular, a transverse ion energy $W_{{\rm i}\perp}$).

In a column of magnetized plasma ($\omega_{\rm ce} = eB/m_{\rm e} \gg \nu_{\rm ei}$, $\omega_{\rm ci} \gg \nu_{\rm ii}$, where $\nu_{\rm ei}$ and $\nu_{\rm ii}$ are the frequencies of electron–ion and ion–ion collisions, respectively), which is the case for separating ICR facilities, electric fields can also be produced directly by end electrodes [1].

The aim of isotopically selective heating is to achieve a considerable difference in the ion temperatures for the selected isotope and for neighboring isotopes. This gives the possibility of separating them by depositing the heated ions on metal plates introduced into the plasma parallel to the lines of force of an external magnetic field and being at a positive electric potential relative to plasma. By choosing a retarding potential V_r, one can control the ratio of heated to cold ion fluxes on a collector, assuring the required separation factor. Since the collectors of heated ions are placed in the plasma, the temperature of the heated ions should not exceed several hundred electron-volts in order to prevent sputtering of the deposit by the heated ions. In the presence of a retarding potential, the heated ions pass to the collector at a lower energy. Nevertheless, it is necessary to prevent sputtering of the deposit on neighboring parts of the collector system.

To accelerate ions to an energy of several hundred electron-volts, it suffices to create in the plasma transverse electric fields with a strength of $0.1-1$ V cm^{-1}, rotating in the sense of the Larmor gyration of ions ($E_{\perp+}$). The transverse energy of the ion that resides in this field for a time t is $W_{i\perp} = E_{\perp+}^2 e^2 t^2/2M$. The time of ion flight across the heating zone in ICR installations is $\tau = L/v_{iz} \sim 10^{-3}$ s. In this time, for example, the energy of the resonance gadolinium isotopic ion increases to 750 eV at $E_{\perp+} = 0.5$ V cm^{-1}.

Isotopically selective heating of ions can only occur at a small relative width of cyclotron resonance lines: $\Delta\omega/\omega_{ci} \ll \Delta M/M$ (here, ΔM is the mass difference for separated isotopes, and M is their average mass). This condition requires first of all high uniformity of the external magnetic field:

$$\frac{\Delta B}{B} \ll \frac{\Delta M}{M} . \tag{1}$$

Inequality (1) should hold over the whole volume of plasma in the heating zone. The required value of the external magnetic field will be discussed below. In advance, we note that in a universal separation ICR installation — that is, one capable of separating isotopes of all chemical elements — the required magnetic field induction is about 4 T.

Since the masses of the isotopes are close, the electric field periodically causes the energy of neighboring isotopic ions to rise as well. The beat period equals $T_b = 2\pi/\Delta\omega$, where $\Delta\omega = \omega_{ci}\,\Delta M/M$ is the difference in the ion cyclotron frequencies for the selected and neighboring isotopes. The distance between the beat nodes on the trajectory of the nonresonance ion is $l_b = 2\pi v_{iz} M/(\omega_{ci}\,\Delta M)$. Thus, for isotope separation it is necessary to use extended heating zones: $L \gg l_b$. Hence, the condition for the selective heating is written down as

$$\frac{2\pi v_{iz}}{L\omega_{ci}} \ll \frac{\Delta M}{M} . \tag{2}$$

Both the conditions for selective ion heating are taken into account in constructing a solenoid, because the dimensions of the domain of the uniform magnetic field cannot be changed after the solenoid has been manufactured. The dimensions of this domain determine the maximal extent of the heating zone L.

These are the necessary external conditions for selective ion heating. The selectivity of heating also depends on internal parameters, namely, ion density and temperature in the plasma. The third condition for selective heating is expressed via the frequency of ion–ion collisions:

$$\frac{\nu_{ii}}{\omega_{ci}} \ll \frac{\Delta M}{M} . \tag{3}$$

The collision frequency is proportional to the ion density: $\nu_{ii} \approx 5 \times 10^{-7} n_i/(T_i^{3/2} A^{1/2})$; here, n_i is the ion number density [cm^{-3}], T_i is the ion temperature in electron-volts, and A is the atomic weight of the element. In gadolinium plasma at $n_i = 10^{12}$ cm^{-3} and $T_i = 5$ eV, we have $\nu_{ii} \approx 3.5 \times 10^3$ s^{-1} ($\omega_{ci} \approx 2.4 \times 10^6$ s^{-1}, $B = 4$ T).

Inequality (3) limits the plasma density only. But due to the Doppler effect the initial temperature of ions should also be limited. The Doppler effect influences the selectivity of heating when the spread of longitudinal ion velocities Δv_{iz} exists:

$$\frac{k_z \Delta v_{iz}}{\omega_{ci}} \ll \frac{\Delta M}{M} . \tag{4}$$

In most plasma sources we have $\Delta v_{iz} \sim v_{iz}$, and for a rough estimate of the broadening of ICR lines it suffices to take the quantity $k_z v_{iz}$, where $v_{iz} \sim (T_{i\|}/M_i)^{1/2}$. Formula (4) illustrates how difficult the selective heating of gadolinium isotopic ions is. At $k_z \approx 0.016$ cm^{-1} (the wavelength is $\lambda = 400$ cm), $v_{iz} \sim 2 \times 10^5$ cm s^{-1} ($T_{i\|} \sim 5$ eV), $\omega_{ci} \approx 2.4 \times 10^6$ s^{-1}, we arrive at $k_z v_{iz}/\omega_{ci} \approx 1.3 \times 10^{-3}$ ($\Delta M/M \approx 6 \times 10^{-3}$).

Let us pay attention to the value of the density of equivalent ion current in the plasma: $g = e n_i v_{iz}$, which at the parameters of the gadolinium plasma considered above amounts approximately to 30 mA cm^{-2}. This value on the order of magnitude equals the ion current in a single beam of an electromagnetic separation facility. Since an ICR installation assumes employment of plasma fluxes with the cross-section area $S \sim 1000$ cm^2, there is an assertion that such a facility can outperform an electromagnetic separator by a factor of 1000 with respect to the separation flux. However, a large electromagnetic separator has more than one beam [16, 17]. In addition, the extraction of a target isotope from a plasma flux is not perfect yet ($0.03-0.3$). Hence, the advantage factor to be spoken about is only several dozen.

After these preliminary remarks, consider the separation process itself. A scheme for ICR separation is actually presented in Ref. [1]. The corresponding constructive design was realized in a TRW patent application [6] (see Fig. 1). For

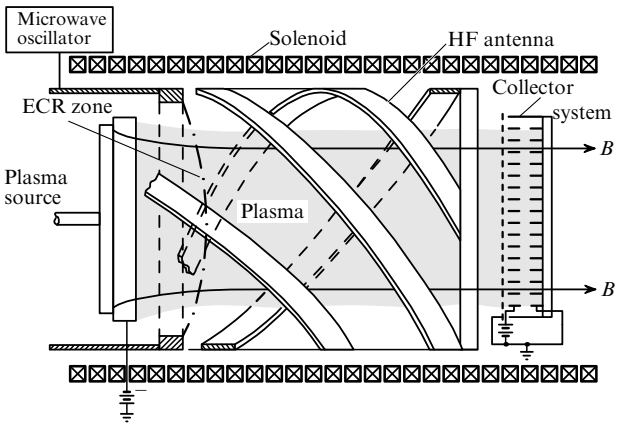

Figure 1. Schematic of an ICR separation facility (patent application [6]).

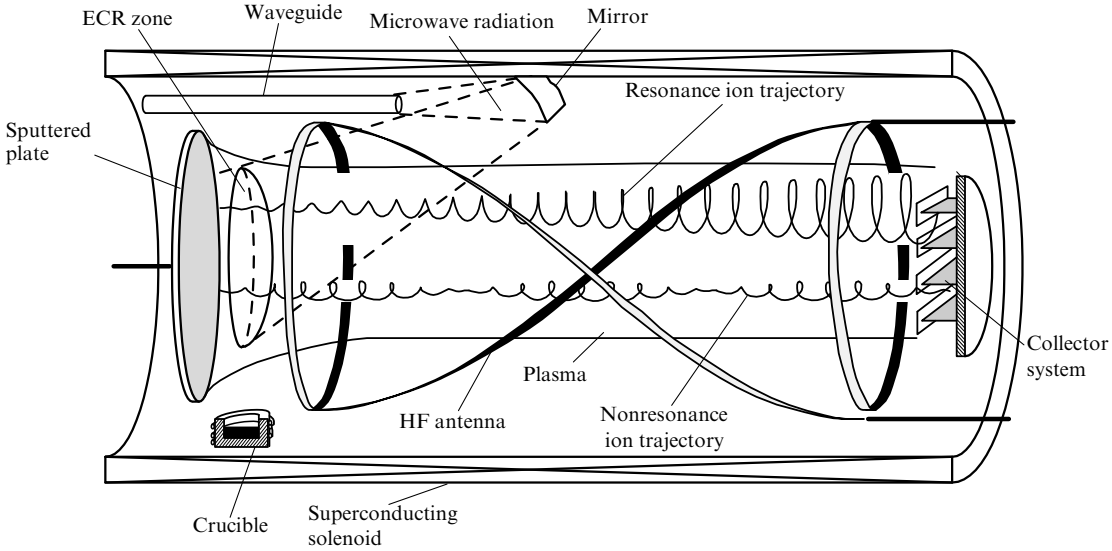

Figure 2. Schematic of the ICR separation process.

describing the separation process, we will also resort to Fig. 2 in which the construction of the experimental setups is shown.

The process starts with the creation of the plasma flow, in which the ion component is formed by ionized atoms of the element whose isotopes are separated. Plasma propagates along a constant magnetic field B. Selective heating occurs in the zone of the uniform field. For producing the electric field $E_{\perp+}$ heating the ions and rotating in the sense of the Larmor gyration of ions, mainly inductive four-phase antennas were employed (see Fig. 1). Their length L varied from a one-half meter to several meters depending on the mass and mass difference of separated isotopes.

Having passed the heating zone, the heated ions are deposited on the plates of a collector system, which are placed parallel to the lines of force of the magnetic field (see Fig. 2). In terms accepted for describing separation facilities, this deposit is called a 'product'. The collector system also serves as a receiver of the plasma stream depleted of target isotopes, i.e. 'waste'. For selecting ions by energies, in addition to applying a positive retarding potential V_r to the plates, a geometrical factor is also used, namely, the gap d between the plates collecting heated ions is chosen close to the double Larmor radius of the heated ions: $d \approx 2r_{Lh}$, where $r_{Lh} = v_{i\perp}/\omega_{ci} = M_i v_{i\perp}/(eB)$, and screens of the height h equal to the Larmor's radius r_{Lc} of cold ions are employed.

The key elements in a separation ICR facility are an extended solenoid, a vacuum system, and a plasma source. An antenna used for selectively heating ions and a collector system are less tedious to fabricate.

A superconductive solenoid for the PPM was created in the USA by General Dynamics Convair Division. The parameters of its cryostat are as follows: the length is 8 m, the outer diameter is above 2 m, the diameter of the internal hole (it is called in Russian papers a 'warm hole' and in English-speaking countries a 'clear bore') is about 1 m. It is known that the weight of the mounted magnet amounts to 44 tons. The cryostat parameters per se characterize the scale of the ICR separation facility. At such dimensions of the clear bore in a magnet, a separation chamber mounted inside it admits a yield of several grams to several dozen grams of product daily, depending on the natural concentration of selected isotope. It was mentioned, however, that the magnetic field in the PPM solenoid is $B \leqslant 1.8$ T, which substantially limits the number of chemical elements whose isotopes may be beneficially separated at this facility.

The solenoid used in the PPM is projected to provide the parameter $\Delta B/B = \pm 0.1\,\%$ at a distance of 5 m. Tests of the solenoid revealed [18] that the uniform zone is approximately 0.5 m shorter. It was evidenced that the magnetic field can be corrected by means of a nonsuperconducting coil placed in the clear bore of the solenoid. There is no information on whether such a correction was used or not in the experiments.

Designers of the solenoid for French facility ERIC, which was initially intended for research purposes (the clear bore diameter is 0.3 m), aimed to provide maximal possibilities for experimenters. With this purpose in mind, windows were made in the magnet for carrying out optical and probe measurements of plasma parameters. The broken homogeneity of the superconducting coil resulted in an enhanced nonuniformity of the magnetic field: at the ERIC facility, the corresponding parameter is $\Delta B/B = \pm 0.5\,\%$. In the MCIRI project (method of cyclotron ion resonance of isotopes) [12], primarily aimed at separating rare-earth isotopes, for which we mainly have $\Delta M/M < 1\,\%$, the solenoid was designed with $|\Delta B/B| \leqslant 3.5 \times 10^{-4}$ at a distance of 4.5 m and a cross-section diameter of 0.4 m.

The requirements of the vacuum system are determined by the necessity to quickly evacuate air from the operation chamber (its volume is several cubic meters) after finishing the mounting work, and to replace the source and collector systems through special locks maintaining the vacuum (vacuum gates with a pass-through diameter of 0.5 m are needed). The ultimate vacuum in the operation chamber should be about $(1-2) \times 10^{-6}$ Torr, which can be provided by oil-diffusion pumps.

A quite perfect method for producing plasma in an ICR installation is atomic ionization by a microwave discharge in the conditions of electron cyclotron resonance (ECR discharge) [19] excited at the electron cyclotron frequency $\omega_{ce} = eB/m_e$. In a magnetic field $B > 1$ T, microwave radiation with a frequency above 28 GHz is needed. Plasma flux with a cross-section area $S = 800$ cm^2 was obtained. In

producing plasma in the source of the ICR facility, it is important to determine the conditions in which the number of multiply charged ions would be minimal because the separation process, naturally, is aimed at obtaining plasma with mainly singly charged ions. In the case where neutral atoms are obtained via target sputtering by ions, a plate made from a metal whose isotopes are separated is placed in a vacuum chamber at the end of the solenoid. The plate is under the negative electric potential $|V| = 2-4$ kV and its near-surface area (see Fig. 2) is irradiated by microwave radiation at a frequency corresponding to the electron cyclotron resonance in the zone several centimeters from the surface (ECR zone).

Initially the ECR discharge is excited in the noble gas filling the chamber. Then the discharge is maintained partially or completely due to the ionization of atoms knocked out from the target. Since the sputtered plate resides at the end of the solenoid, where the magnetic field is weak, the electron component of produced plasma is trapped between the negatively charged plate being sputtered and the magnetic mirror formed by the magnetic field that increases in the opposite direction. The property of the increasing magnetic field to reflect charged particles is explained by the fact that the quantity $W_\perp/B$ is an adiabatic invariant of particle motion.

Going away of plasma from the source into the zone of high-frequency ion heating starts with the escape of electrons (which are faster particles). The magnetic mirror passes those charged particles that have at the initial point of trajectory an angle between the velocity vector and the lines of force of the magnetic field less than the angle φ_0 determined by the relationship $\sin\varphi_0 = (B_0/B_\mathrm{m})^{1/2}$; here, B_0 is the magnetic field at the point where a neutral particle ionizes, and B_m is the maximal field in the magnetic mirror. The ratio B_m/B_0 is called the mirror ratio, and the trap for charged particles made by two magnetic mirrors is called a magnetic mirror trap. The cone of directions of free escape for charged particles from the magnetic mirror trap is called the loss cone.

Then, the electron and ion fluxes level off. It is assumed that along a uniform magnetic field plasma moves at the ion-sound speed $v_z = [(T_\mathrm{e} + T_\mathrm{i})/M_\mathrm{i}]^{1/2}$.

An electron trap is also expedient in cases where neutral particles are obtained via thermal evaporation (a crucible source of atomic vapor [20]). To this end, a metal plate should be placed at a negative potential of several dozen volts near the ECR zone, which does not cause sputtering of the plate (Babichev et al. [21]).

A gyrotron is a generator of microwave radiation, which is suitable for an industrial facility, i.e., capable of providing the equivalent ion current of several dozen amperes. In Ref. [19], the continuous-wave gyrotron VGA-8000 (28 GHz, 100 kW) of the Varian Co. was used. In producing the metal plasma flux, to a 1 A equivalent ion current, the corresponding microwave power consumed ranges 0.5–1.0 kW [22]. At the French SuperERIC facility [23] the stationary gyrotron Bylina (37.5 GHz, 15 kW) manufactured at the production association Salyut in Nizhny Novgorod, Russia was used.

A less powerful ECR discharge in a magnetic mirror trap was utilized in the sources of multiply charged ions for accelerators [24], where it displaced other types of discharges. The old question of whether other types of electric discharges can be applied in a source of a separation ICR facility [25] is still topical. Gyrotrons are expensive and have a short operation lifespan. Employment of a microwave source is hindered by the necessity of protecting the entrance windows for microwave radiation, waveguides, and microwave mirrors (see Fig. 2) against dusting by a working matter.

Among plasma accelerators (see the monograph by Morozov [26]) there is no ready construction suitable for producing a wide plasma flux (~ 1000 cm^2) in strong magnetic fields. An alternative approach to the problem of producing metal plasma at an ICR facility is outlined in work carried out at the Budker Institute of Nuclear Physics, Siberian Branch of RAS (Volosov et al. [27]). The discharge configuration was studied with radial electric and axial magnetic fields in a coaxial axially symmetric system comprising a rod cathode on an axis and a cylindrical anode. The cathodes were made from titanium and molybdenum and in practical employment of the source they should be made from metal elements whose isotopes are separated. The discharge ignition was initiated by pulse hydrogen puffing. Then, the discharge was maintained due to the ionization of atoms obtained in cathode sputtering under the action of accelerated ions. Presently, only a quasistationary discharge has been obtained (0.25 s). The equivalent ion current in plasma flux through the output hole of the source was about 1 A (50 mA cm^{-2}).

Numerous works confirm the efficient ionization of gases by helicons (waves with circular polarization in the frequency range from ω_ci to ω_ce, propagating in plasma along the magnetic field). Oscillations were mainly excited at frequencies close to the geometrical mean of the ion and electron cyclotron frequencies: $\omega = (\omega_\mathrm{ci}\omega_\mathrm{ce})^{1/2}$ [28]. The excitation technique at these frequencies is simpler than that at the ECR frequency. Nevertheless, the possibility of gas ionization, rather than of atomic vapor, was demonstrated. The authors of Ref. [29] have probably carried out experiments on ionizing atomic vapor; however, they did not describe them. Most experiments on gas ionization have been carried out in weak magnetic fields ($B < 0.2$ T).

Thus, in a large separation ICR facility there is no alternative to the ECR plasma source.

In describing the process of ICR separation, we should also demonstrate the importance of the constructions of the HF antenna and collector system, which we will do in what follows.

3. Selectivity of the separation method based on ion cyclotron resonance (ICR)

Selective heating of isotopic ions necessitates that the frequency interval between resonance lines, $\Delta\omega = \omega_\mathrm{ci}\Delta M/M$, be greater than the line width which in collisionless plasma is defined by the formula

$$\Delta\omega_\mathrm{D} = \Delta(k_z v_z) = \Delta v_z k_z + \Delta k_z v_z \approx \left(\frac{T_\mathrm{e}}{M_\mathrm{i}}\right)^{1/2} (k_{z0} + \Delta k_z)\,.$$

Here, k_{z0} corresponds to the maximum of the k_z-spectrum of an HF electric field in a plasma, and Δk_z is its width. It is assumed that the spread of ion longitudinal velocities in the plasma flux equals $\Delta v_{\mathrm{i}z} \sim v_{\mathrm{i}z}$ and $T_\mathrm{e} > T_\mathrm{i}$. In this form the quantity $\Delta\omega_\mathrm{D}$ is presented in the work by Compant La Fontaine and Pashkovsky [30]. The authors [30] call it Doppler broadening and do not use a particular term for the broadening connected with a finite time of ion flight through a heating zone [see Eqn (2)], which is included in the second

summand. The velocity spread of ions produced by a plasma source cannot actually be changed. The problem of tapering the k_z-spectrum of the electric field $E_{\perp+}$ heating the ions was mainly discussed. The penetration of left-handed polarized electric field component E_+ into the magnetized plasma at the ICR frequency is related to the quantity k_{z0} as well. In choosing the induction antennas for exciting an electric field in a PPM plasma, an effort was made to meet the necessary condition $k_{z0} \neq 0$ [31]. Spiral antennas were actually used, which created fields at a wavelength equal to or twice the antenna length: λ- and $\lambda/2$-antennas. The work by Ustinov [32] was devoted to problems concerning electric field excitation in a plasma of a large ICR separation installation.

The range of resonance frequencies for ions of chemical elements whose isotopes are appropriate for selection using the ICR approach covers 0.1–1 MHz. The amplitude of the electric field in the plasma should be within the range of 0.1–1 V cm^{-1} (see Section 2). Calculations demonstrate that the rise in current amplitude in a spiral multiphase antenna with the aim of exciting stronger electric fields may be inefficient. The antennas influence ion motion along the basic magnetic field, accelerating or decelerating them, depending on the spiral twist direction, which disturbs the cyclotron heating conditions chosen initially (Laz'ko [33]).

Similarly to other separation methods, the selectivity of the ICR method is estimated finally by a mass spectrometric analysis of the isotope composition of enriched samples (MS analysis). The isotopic selectivity of cyclotron heating can be preliminarily estimated by means of an electrostatic analyzer of ion energies. In Refs [1, 20, 34], an open analyzer of ion transverse energies was used [35], introduced in the cross-sectional plane of the plasma flux and consisting of a metal tube (screen) and plane ion collector placed inside it. The collector went deep into the tube at a distance of several dozen electron Larmor radii, so that theoretically it could detect the ion flux only. Actually, electrons reached the collector. Later we will refer to this fact. To resolve ICR lines, it was necessary to maintain a positive electrical potential V_r at the collector. It should be noted that the operational principle of this analyzer is explicitly used in the construction of product collectors at separation ICR facilities [6].

In Fig. 3, a resonance curve obtained in the course of tuning the frequency of the heating field in the range that includes the resonance frequencies of all six isotopic ions of calcium is presented [20]. The main specificity of this spectrum is the line of the $^{48}Ca^+$ ion, which is observed even though the natural concentration of this isotope constitutes only $\approx 0.2\%$. The fact that ions of isotopes with lower concentration are heated more efficiently was often observed, starting from the experiments on the selective heating of potassium ions in a potassium plasma ($^{39}K^+$ and $^{41}K^+$ [1]).

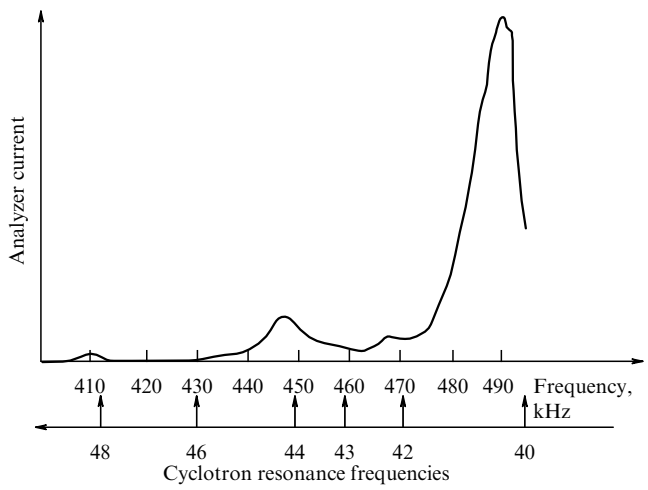

Figure 3. ICR spectrum of calcium isotopic ions in a calcium plasma ($B = 1.287$ T, $V_r = 50$ V).

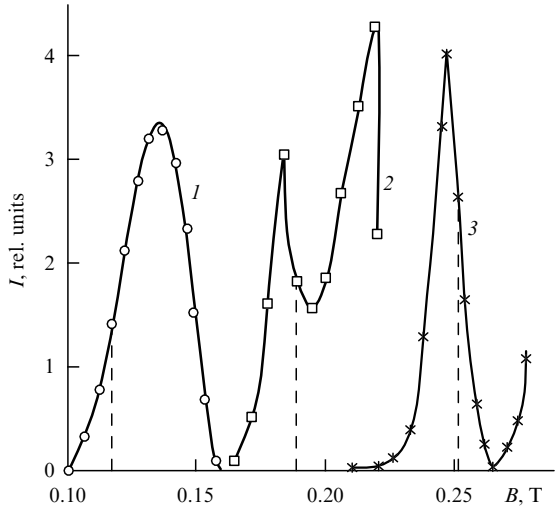

Figure 4. Ion current to the collector of an analyzer in a lithium plasma versus a magnetic field induction at various frequencies of HF heating: *1*—$f = 300$ kHz, *2*—$f = 480$ kHz, and *3*—$f = 640$ kHz. Dashed lines indicate the values of resonance fields for the $^6Li^+$ ion.

The determining effect on the selectivity of ion heating is produced by a magnetic field — the spacing between the resonance lines rises proportionally to its magnitude: $\Delta\omega \sim B$. In Fig. 4, analyzer-collector ion currents in lithium plasma are depicted [8]. A closed multigrid analyzer found application in work [8] in which, while ions are detected, electrons conventionally do not reach the collector.

The results of mass spectroscopic (MS) analysis of enriched material samples can be characterized by the separation factor. In contrast to industrial methods, in the developed approaches to separation it is customary to calculate the separation factor relative to the natural composition rather than to isotopic concentration in a waste: $\alpha = C_p(1 - C_0)/C_0(1 - C_p)$. Here, C_p is the concentration of the enriched isotope in the product, and C_0 is its feed concentration. The formula refers to binary isotopic mixture. In the case of a multicomponent mixture, terms $(1 - C_0)$ and $(1 - C_p)$ refer to the total concentration of all the remaining isotopes. The depletion coefficient for plasma flux with respect to the separated isotope is calculated from the concentration of this isotope in a waste: $\beta = C_w(1 - C_0)/C_0(1 - C_w)$. Since extraction of a target isotope from a plasma flux using the ICR method is not perfect, we have $1/\beta < \alpha$. A minor part of the target isotope quantity carried by the plasma flux is found in the product. The greater part of the target isotope flux becomes waste, which is thus depleted to less a degree than the product is enriched. Due to this circumstance, the question of removing a certain isotope from the isotopic mixture has not been posed for the ICR method.

Table 1. Results of enrichment of eight chemical elements with certain isotopes (ERIC facility [23]).

Resonance isotope	Natural concentration, %	Relative mass difference, %	Magnetic field induction, T	α	β
^{43}Ca	0.134	6.86	0.714	11.8 (31*)	0.246
^{44}Ca	2.06	9.05	1.29	18	0.174
^{48}Ca	0.186	16.5	1.29	10	0.108
^{48}Ca	0.186	16.5	2.07	24 (30*)	—
^{48}Ca	0.186	16.5	0.723	24 (133*)	0.335
^{64}Zn**	49.8	4.44	0.716	1.04	0.263
^{68}Zn	18	4.95	1.25	4.42	0.63
^{110}Cd	12.4	2.74	1.29	2.65	0.62
^{132}Ba	0.104	4.11	1.29	8.5	0.76
^{176}Yb	12.8	1.89	2.94	6	0.92
^{50}Cr	4.35	4.3	1.54	13.6*	—
^{62}Ni	3.35	5.49	0.95	3.28	0.96
^{102}Pd	1.03	4.47	1.714	2.04	0.94

* On part of the collector plate only.
** Isotope removal from natural composition.

Extraction is characterized by the extraction coefficient γ which is determined as the ratio of the target isotope quantity on the product collector to that transferred by the plasma flux during the extraction process.

Moving magnetized two-component plasma is specific in that a deposit on the surface of any plate or cylinder placed in the plasma parallel to the external magnetic field will be enriched with the heated component even without a potential V_r. The heated particles pass to the plate or cylinder from a layer of thickness $2r_{Lh}$, while the cold particles do this from a layer of thickness $2r_{Lc}$, hence the separation factor is $\alpha \approx r_{Lh}/r_{Lc}$. This factor is called 'geometrical' [22]. In the presence of the retarding potential V_r, the separation factor is higher; however, the quantity of extracted target isotopes usually reduces.

Enriched potassium samples were the first obtained by the ICR method [1]. MS analysis was performed of the potassium deposit on a tungsten strip placed in a plasma behind the zone of selective heating and parallel to the lines of force of the magnetic field. In heating $^{41}K^+$ ions, the separation factor was $\alpha \approx 60$. In heating $^{39}K^+$ ions, it was $\alpha \approx 3.5$. A positive retarding potential $V_r = 4$ V was applied to the strip. In enriching nickel with the ^{58}Ni isotope at large facilities [4], its concentration increased from 68% to 97% ($\alpha \approx 15$), and in enriching it with the ^{62}Ni isotope [36], the increase went from 3.66% to 40% ($\alpha \approx 17.5$).

Experiments on isotopic enrichment of ten chemical elements were carried out at the French facility ERIC. A part of the results is presented in Table 1 [23]. Data on magnesium and gadolinium are given in Ref. [37] and Ref. [38], respectively. In addition, conditions of an experiment on enriching samples with Mg, Ca, Zn, Ba, and Yb isotopes are presented in Ref. [37]. Employment of laser-induced fluorescence allowed the authors to measure the temperature rise for nonresonance particles (the temperature of $^{138}Ba^+$ ions increased from 0.5 eV to 3.5 eV as $^{132}Ba^+$ ions were heated), i.e., to indicate a reason for selectivity limitation. Agreement was found between the results of some experiments and the selective heating theory by Compant La Fontaine and Pashkovsky [30]. In particular, the broadening and shift of the resonance line for the $^{44}Ca^+$ ion in calcium plasma corresponded to the theory (see Fig. 5).

The data presented are insufficient for a complete analysis of the separation process at ERIC , because the flux of matter to the product collector was not specified. It was only mentioned that the flux amounted to several hundred milligrams per hour at the feed flux in the range from 10 to 80 g per hour.

Data on ^{48}Ca, ^{44}Ca, ^{132}Ba, and ^{176}Yb from Table 1 show a reduction in the separation factor α at a lower ratio $\Delta M/M$. The less the ratio for a separated isotope, the more difficult it is to meet conditions (1) – (4) for isotopically selective heating of its ions. Nevertheless, the experiments at the French facility did not evidence a dependence of the separation effect on the magnetic field strength. This can only be at a higher inhomogeneity of the magnetic field than the value of 1% given by the authors [23]. Selective heating necessitates the fulfillment of condition (1). This condition held in the experiments with all the above-mentioned elements except for gadolinium. Nevertheless, no dependence of α on the field strength was observed, even in the case of calcium enrichment with the ^{48}Ca isotope. This fact, naturally, did not prevent the authors of Ref. [23] from considering an increase in the magnetic field intensity as a method for enhancing selectivity.

In Table 1, a unique result is presented (see the third row): in enriching calcium with the ^{48}Ca isotope, its concentration in the waste falls by an order of magnitude ($\alpha\beta \approx 1$). The

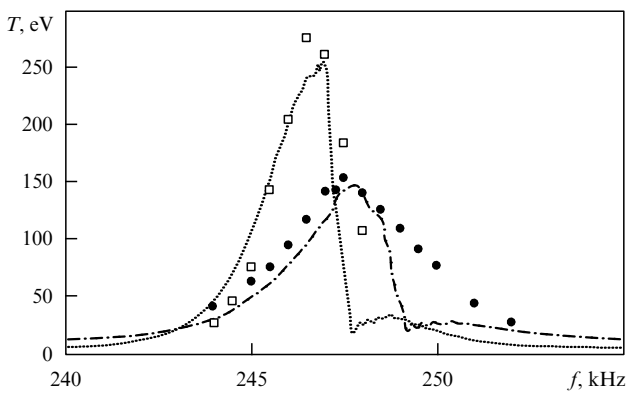

Figure 5. The transverse temperature of $^{44}Ca^+$ ions in a calcium plasma versus the frequency f of the heating field. Squares correspond to $n = 4.2 \times 10^{11}$ cm^{-3}, $T_e = 1.2$ eV, $T_{i0} = 4.0$ eV; circles correspond to $n = 0.9 \times 10^{11}$ cm^{-3}, $T_e = 2.8$ eV, $T_{i0} = 10.0$ eV. The curves show calculated results from Ref. [30].

result on nickel enrichment with the ^{62}Ni isotope ($\alpha \approx 3.3$) is very poor compared to Ref. [36].

Generally, taking into account the experiments on separating lithium isotopes ($\alpha = 76$) [39], one may conclude that the separation factor α at ICR facilities falls in the range from several units to several dozen. The record high value $\alpha = 133$ refers to calcium enrichment with the ^{48}Ca isotope (see Table 1). Minimal values ($\alpha \approx 2-5$) refer to gadolinium enrichment with the ^{157}Gd isotope [14, 36].

In all isotope separation methods, including the ICR approach, the selectivity falls at greater separated fluxes. Hence, the parameter α does not completely characterize the separation method if the product flux is not specified. There are three works mentioned above in which both quantities were measured [14, 36, 39]. Section 4 is devoted to these works.

4. Productivity of ICR facilities

To date, the accumulation of product at ICR facilities, in contrast to electromagnetic separators, can be monitored at $V_r = 0$ only. The reason has to do with the appearance and even predominance of an electron current to the collector in the presence of a retarding potential V_r. In Fig. 6, electric characteristics are given for a collector system employed in the ICR separation of lithium isotopes [40]. The formula defining the product quantity in an electromagnetic separator has the form [16, 17] $Q \approx 3.7 \times 10^{-2}\, AC_0\, I$ [g h^{-1}], where A is the atomic weight of the element, C_0 is the fraction of the separated isotope in the initial mixture, and I is the total collector ion current in amperes. For the ICR method, this formula is useful in that it allows one to solve an inverse problem, namely, to determine the collector ion current from the quantity of product.

It was mentioned above that the construction of a product collector is similar to that of an electrostatic analyzer [35]. In an analyzer, the collector electron current could be noticeably reduced by additionally applying a potential slightly exceeding V_r by a fraction of a volt to the tube. No such experiments have been carried out with collector systems of ICR facilities. So far, the mass of collected material (enriched and depleted) can only be determined by removing the collector system from the installation.

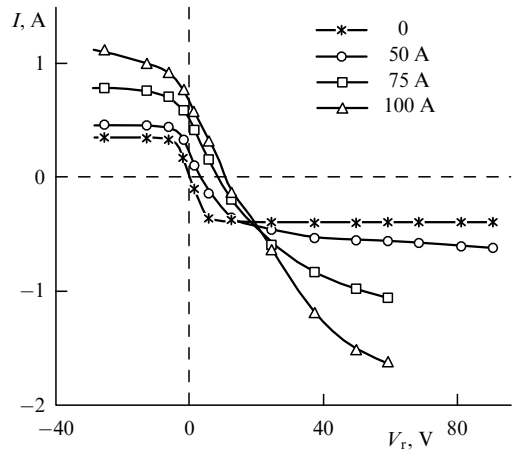

Figure 6. Current–voltage characteristics for a collector of products in a lithium plasma at various currents in an HF antenna.

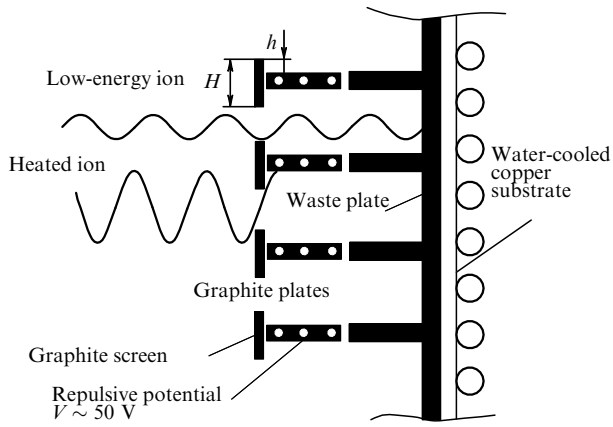

Figure 7. Schematic of a traditional ICR collector system.

In Fig. 7, a traditional collector system for an ICR facility is presented in more detail than in Fig. 1. This construction was used in experiments on separating gadolinium, dysprosium, and erbium isotopes [14], the results of which will be given below. Now consider other specific features of operation for such collector systems (the first feature already mentioned is the predominance of electron current to the product collector).

Let us next note the reduction in the deposit surface density and target isotope concentration at the collector as one recedes from the screen along the magnetic field B [41] (see Fig. 8). As a rule, the deposit spreads from the start of the collector plate to a distance equal to the step of the ion spiral path in the cross section of plasma flux, where the extraction occurs. Finally, there is an unavoidable deposition of material on the screens themselves, which reduces the separation flux by approximately the value of the ratio $H/2r_{\mathrm{Lh}}$ (H is the screen width). This correction is usually neglected and the extraction factor γ for the target isotope accounts for both the physical processes and these obvious losses. The quantity γ is also affected by the screen thickness because some heated ions do not reach the collector and deposit onto a lateral surface of the screen. The separation factor α depends on the screen height h (see Fig. 7). Under a long exposure of the collector system to a plasma flux, the deposit changes the screen thickness and height, which influences the product quality. In addition, the elements of the collector system should be cooled if thermal fluxes are strong and may cause evaporation of the deposits. If neutral particles in the plasma source are produced by sputtering of the metal plate, an additional thermal load on the screens and waste plates arises due to an exposure to accelerated electrons (ion–electron emission in sputtering).

The fall in concentration along the collector plates means that not the entire collected mass is actually the product. A part of the deposit may not possess the required enrichment. In this case, the extraction factor γ is calculated by a sufficiently enriched part of the deposit. Works by Karchevskii and Potanin [42, 43] are devoted to the properties of collector systems and the dependence of the extraction factor on the initial shape of the ion distribution function over energy.

To characterize the separation process itself, it is reasonable to estimate the productivity of ICR facilities by the results of the continuous stage of operation. The product

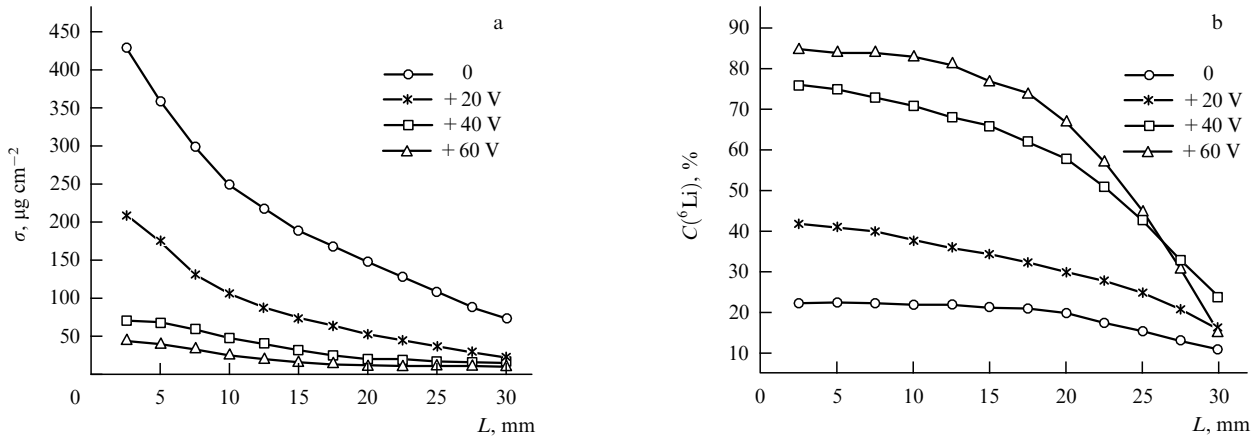

Figure 8. Distributions (a) of the deposit surface density on a collector for enriched lithium, and (b) ^{6}Li isotope concentration in the deposit at various V_r. The diagnostic collector was a cooled copper plate 30×10 mm^2 in area.

quantity produced daily or, even more so, weekly depends on the facility's technical level (the presence of vacuum gates, time needed to change the source of plasma and collector, etc.).

The experiment on nickel enrichment by the ^{62}Ni isotope, carried out at TRW [36], is to date the closest to an industrial scale. The equivalent ion current I_{equ} in plasma flux was above 64 A (the intensity of ion flux was above 5×10^{17} cm^{-2} s^{-1}, and $S = 800$ cm^2). The extraction factor was $\gamma \geqslant 0.15$. For a week, the daily production of nickel enriched to 40% ($\alpha \approx 17.5$) with the ^{62}Ni isotope was 13 g. The experiments were conducted for only part of the day: they were interrupted because the screen dimensions noticeably changed due to nickel deposition. It seems likely that the productivity reached 2 g h^{-1}, since the collector current for the ^{62}Ni isotope was $i \geqslant I_{equ}\, C_0 \gamma \geqslant 0.35$ A.

Let us next consider the results of the experiment on enriching lithium with the ^{6}Li isotope, performed at the Institute of Molecular Physics of the Russian Research Centre 'Kurchatov Institute' at the Siren' installation [39]. Enriched lithium collector deposits were compared for those cases when a collector system (CS) was placed in a uniform magnetic field and in the domain of diverging lines of force near the end of a solenoid. The possibility of a longer CS exposure in a weaker magnetic field was studied. We present below the results obtained in the uniform magnetic field only.

The equivalent current of lithium ions in the plasma flux was 5–10 A. Two CSs were employed. In the first one, enriched lithium samples were produced on two orthogonal copper plates with a width of 60 mm, length of 50 mm, and thickness of 4 mm. The plates had copper screens with $h = 2$ mm, $H = 8$ mm, and a thickness of 3 mm. In the second CS, the enriched matter was collected on the external surface of a cylinder 16 mm in diameter and from the internal surface of a thin-walled hollow cylinder with a diameter of 64 mm. The hollow cylinder was placed with a 45-mm shift along the plasma flux. The cylinders were closed from the directed plasma flux by a disk screen 20 mm in diameter ($h = 2$ mm) and an annular screen with an internal diameter of 60 mm and external diameter of 75 mm ($H = 7.5$ mm, $h = 2$ mm), respectively. The collectors were under the same retarding potential V_r. The construction of the cylindrical CS is given in report [44].

The extraction lasted for three minutes. From the deposit mass measurements at the collector and from MS analysis, the separation factor α and extraction factor γ were calculated for the product parts with different concentrations of the ^{6}Li isotope. The feed concentration of the ^{6}Li isotope reached 7.5%. These results are presented in Tables 2 and 3. The lithium mass is given in milligrams.

It should be explained that the difference in α and γ values in Tables 2 and 3 does not testify to the principal advantages

Table 2. Separation factor α, concentration $c(^6\text{Li})$ of ^{6}Li in deposit fractions, deposit mass m in fractions, and extraction factor $\gamma(^6\text{Li})$ from plasma flux. A plane collector for enriched lithium, $V_r = 60$ V.

Fractions	$m\,(^6\text{Li})$	$m\,(^7\text{Li})$	$c\,(^6\text{Li})$	α	$\gamma\,(^6\text{Li})$
Screens	3.769	41.101	0.084		0.482
$c > 0.2$	1.759	1.839	0.489	11.8	0.225
$c > 0.5$	1.157	0.717	0.617	20	0.148
$c > 0.6$	0.818	0.444	0.648	22.7	0.105
$c > 0.65$	0.404	0.209	0.658	23.7	0.052
Collector	1.957	3.562	0.354	6.8	0.250
Waste	2.094	61.372	0.033		0.268
Total	7.820	106.035	0.069		1.000

Table 3. Separation factor α, concentration $c(^6\text{Li})$ of ^{6}Li in deposit fractions, deposit mass m in fractions, and extraction factor $\gamma(^6\text{Li})$ from plasma flux. A cylindrical collector for enriched lithium, $V_r = 60$ V.

Fractions	$m\,(^6\text{Li})$	$m\,(^7\text{Li})$	$c\,(^6\text{Li})$	α	$\gamma\,(^6\text{Li})$
Screens	3.941	47.187	0.077		0.441
$c > 0.2$	0.899	0.586	0.605	19	0.101
$c > 0.4$	0.794	0.348	0.695	28	0.089
$c > 0.6$	0.557	0.113	0.831	61	0.062
$c > 0.8$	0.375	0.067	0.849	69	0.042
$c > 0.83$	0.253	0.041	0.860	76	0.028
Collector	1.014	1.550	0.395	8	0.113
Waste	3.984	67.900	0.055		0.446
Total	8.939	116.637	0.071		1.000

of one CS over the other. Both plane and cylindrical CSs yield a product of similar quality with the same extraction factor if the dimensions are chosen properly, i.e., in accordance with the parameter r_L and radius of plasma flux. A drawback of these CSs is an approximately 40% loss of substance on the screens. However, at this stage of the investigation the problem of improving the collector systems had not yet arisen.

Pay attention to the string 'collector'. The lithium mass collected on the plane collector was 5.52 mg with $c(^6\mathrm{Li}) \approx 35\,\%$, $\alpha = 6.8$, $\gamma = 0.25$ (see Table 2), and it was 2.56 mg with $c(^6\mathrm{Li}) \approx 40\,\%$, $\alpha = 8$, $\gamma = 0.11$ (see Table 3) on both cylindrical collectors. The ion currents to the collectors can be estimated from the quantity of the collected substance, which gives approximately 0.4 A and 0.2 A, respectively. Actually, an electron current was measured, which was several amperes. The masses of more enriched lithium on the collectors were approximately the same, namely, 1.262 mg [$c(^6\mathrm{Li}) \approx 65\%$ for the plane collector] and 1.142 mg [$c(^6\mathrm{Li}) \approx 70\%$ for the cylindrical collectors]. In reality there was no lithium with the isotope concentration $c(^6\mathrm{Li}) > 66\%$ on the plane collector. Thus, the productivity of a facility with these collector systems for lithium enrichment (65–70%) with the isotope ^{6}Li may attain ≈ 20 mg h^{-1}.

The experiments on gadolinium, dysprosium, and erbium isotope separation carried out at TGX [14] were not as successful as those on nickel isotope separation performed at TRW [36] at the same facility. The results of some experiments were given as an example. In addition, a summary table was presented, according to which the fluxes of enriched gadolinium (36% ^{157}Gd), dysprosium (67.5% ^{164}Dy), and erbium (37.5% ^{167}Er) were 0.72, 0.86, and 0.92 g h^{-1}, respectively. The extraction factors γ were 0.03, 0.04, and 0.04, respectively. The feed concentrations of these isotopes were 15.7% for ^{157}Gd, 28.2% for ^{164}Dy, and 22.9% for ^{167}Er. Data on gadolinium were based on the results obtained with a diagnostic collector only. In the experiments with dysprosium and erbium, full-scale collectors were employed as well.

Equivalent ion currents in Gd, Dy, and Er plasma fluxes were not specified. However, they can be estimated based on the substance fluxes to the product collector and extraction factors presented. For I_{equ} this gives ~ 0.6 of the rectifier load currents through sputtered targets in the plasma source, which yields 9.5 A (Gd), 8.5 A (Dy), and 6.0 A (Er). The above-given values of the fluxes to collectors for enriched gadolinium, dysprosium, and erbium correspond to the ion currents of 0.12 A, 0.14 A, and 0.15 A, respectively.

The last experiments are specific, first and foremost, in the low selectivity of the separation process. The separation factor α was 3.0 for Gd, 5.3 for Dy, and 2.0 for Er. Such a result was not unexpected, either for the authors or for other researchers. The minimal mass difference $\Delta M = 1$ between the selected isotope and neighboring isotopes and the high concentration of the latter (Gd, Er) pointed to the complexity of the problem in advance. A higher separation factor in the ^{164}Dy isotope extraction can also be explained: this is an extreme isotope with a single neighbor (^{163}Dy).

Prior to the start of experiments, the results of preliminary calculations were published [13] concerning the quality and quantity of the product expected. Gadolinium with the required quality $c(^{157}\mathrm{Cd}) > 70\%$ might be obtained according to calculations in the magnetic field with an induction of 6 T. In the actual field of this installation (1.8 T) according to the same calculations the maximal separation factor for gadolinium isotopes was $\alpha \approx 4.5$. In a particular experiment with gadolinium [14], the calculated value was reached for this parameter: $\alpha \approx 5$. Recall that ISTC project 0830 [12] assumed the creation of a solenoid with a maximum magnetic induction of 4 T for an ICR facility.

The quantity of obtained enriched material was an order of magnitude less than the calculated value, which was unexpected even for the authors. Analysis of the experiments leads one to conclude that this fact is explained by a low temperature of ions in a plasma produced by a source. In order to meet the necessary condition for the selectivity of heating, $\nu_{\mathrm{ii}}/\omega_{\mathrm{ci}} \ll \Delta M_{\mathrm{i}}/M_{\mathrm{i}}$ (see Section 2), and reach a separation factor of several units, the experimenters reduced the plasma density. Obviously, these experiments on separating isotopes of rare-earth elements were carried out at the plasma number density $n_{\mathrm{i}} < 10^{12}$ cm^{-3} (according to the parameter I_{equ}, we have $n_{\mathrm{i}} v_{\mathrm{i}} \leqslant 7.5 \times 10^{16}$ cm^{-2} s^{-1}). Correspondingly, the flux of matter to the product collector was also reduced.

One might think that neutral particles ionized in a plasma source due to sputtering of a metal target guarantee obtaining a sufficiently high ion temperature because relatively fast neutral particles are ionized [45]. The initial temperature of gadolinium ions was measured earlier at the French facility ERIC [38] and ranged 2–3 eV. Unfortunately, these data were not confirmed in the publication [46] that followed the report. Seemingly, there were doubts in the results of ion temperature measurements. In these experiments, the operation mode of the plasma source was not arduous: $I_{\mathrm{equ}} < 10$ A, and the temperature of the gadolinium ions was probably the same. Nevertheless, we may use the ion temperature measured in Ref. [38] for estimating ν_{ii}: the average temperature is $T_{\mathrm{i}} = 2.5$ eV, then at $n_{\mathrm{i}} = 10^{12}$ cm^{-3} we have $\nu_{\mathrm{ii}} \approx 5\times\ 10^{-7}\, n_{\mathrm{i}}/T_{\mathrm{i}}^{3/2} A^{1/2} \approx 10^4$ Hz ($\omega_{\mathrm{ci}} \approx 1.1\times 10^6$ s^{-1}). Indeed, at the given plasma density, collisions prevent gadolinium isotopic ions from selective cyclotron heating ($\Delta M/M \approx 0.006$).

In the projects concerning a practical implementation of the ICR method, the initial temperature of the ions was taken in the range of 5–10 eV [47, 48], hence the influence of ion–ion collisions was neglected. It was assumed that in using cathode sputtering in the plasma source the necessary initial ion temperature is obtained automatically, whereas thermal evaporation of a working agent may necessitate additional plasma heating. It follows from report [14] that in separating the isotopes of rare-earth elements, cathode sputtering in the plasma source cannot provide the necessary initial temperature of ions. Variants of designing an efficient source of gadolinium plasma are in general based on evaporation [49, 50]. Thus, the search for methods for preliminary plasma heating to the temperature of 5–10 eV is becoming urgent.

The experimental results considered above give a direction for the further development of the ICR isotope separation method. Now, the most important steps towards isotope separation of rare-earth elements seem to be increasing the magnetic field and initial temperature of the ions. The extraction factor γ has the sense of the part of heated ions relative to their total number for a selected isotope. In cold and dense plasma, selective heating is only possible for particles from the 'tail' of the ion energy distribution. Starting from paper [1], the assumption is made that the runaway effect may favor heating most of the ions. The corresponding threshold value is $E_+ > 10^{-12} n/T_{\mathrm{i}}$ [V cm^{-1}]

[51], where T_i is taken in electron-volts. One can see that the threshold fits the interval of excited electric fields (see Section 3). However, calculation of the time of ion passage to the continuous acceleration mode in fields of such intensity has not yet been performed. This time should be shorter than the time $\tau = L/v_{iz}$ of ion flight through the cyclotron heating zone. We may assume that in experiments on ICR separation the ion runaway effect has not yet been observed. There are no theoretical works on this problem either.

5. Patents and projects

We can only list here part of the numerous suggestions on enhancing the ICR method for isotope separation. Most of them have not been verified yet due to the limited experimental base.

In the source for gadolinium plasma, it was suggested to use thermal evaporation instead of sputtering (Louvet [49]). In order to create an axially symmetrical plasma flux [the evaporator is usually placed at the periphery (see Fig. 2), hence, the plasma density falls along the diameter of a horizontal plasma column] it was suggested to mount a solenoid and vacuum chamber vertically. The vertical position of the installation would result in additional difficulties in the operation and in carrying out experiments. The reason for abandoning cathode sputtering in creating a gadolinium plasma or a plasma of other rare-earth elements is substantial: these metals possess low thermal conductivity, and sufficiently thick targets (with a thickness of greater than 10 mm) cannot be cooled by a contact method in the case of an intensive separation process. A radical solution to the problem, suggested by Karchevskii and Potanin [50], was to employ a moving target (a rotating disk) and radiative cooling. The authors also suggested electron-beam evaporation of gadolinium and other refractory metals.

Various suggestions are presented in patent [52] (Wong, Rosenthal), but the main one defines concretely an early proposition by Dawson [53] to employ a magnetic mirror for separating heated and cold ions. The authors of patent [52] realized their ideas in separating rubidium isotopes [10]; however, only preliminary results are available. Employment of a magnetic mirror for plasma separation process will only be efficient in those installations with a small cross section of plasma flux since only a peripheral annular collector can be used for depositing selectively heated ions reflected from the magnetic mirror. Dawson is sometimes referred to as the inventor of the ICR method for isotope separation and the separation process is called the Dawson process, rather than PSP.

Consider the choice of an HF antenna for selective ion heating in a plasma. Volosov et al. [54–57] do not rely on the employment of a screw antenna (shown in Figs 1 and 2) in separating gadolinium isotopes (plasma sources developed by these authors were presented in Section 2). Indeed, in contrast to screw antennas, the solenoid antenna recommended by the authors of Refs [54–57] produces fields possessing a narrower spectrum with respect to longitudinal wavenumber k_z. But in this case, the radial distribution of the electric field excited in the plasma is not acceptable, because the field is absent in the paraxial domain of the plasma column. In the last project [56, 57], the authors do not use a solenoidal antenna and plan to employ an electrode-capacitance antenna. We cannot agree with such a choice: electrode-capacitance antennas based on the property of magnetized plasma to transfer the potential of end electrodes along the plasma column create electric fields with $k_z \approx 0$, which are shielded at the resonance particle density of $10^9 - 10^{10}$ cm^{-3} [58–60]. This is the main hindrance for practical employment of contact methods for exciting electric fields. Similar speculations are given in Ref. [57], but the authors see a solution in modulating the shape of the HF voltage applied to the electrodes. This proposition, however, has not been described.

The work by Timofeev [61] is devoted to a comparison of various HF antennas employed in ICR separation of isotopes. The author suggested a new antenna (he calls it a 'spiral antenna'), which outperforms screw and solenoidal antennas by a characteristic collection (the width of the k_z-spectrum, the possibility of exciting the $m = 1$ mode, the Q-factor). In addition, a new approach is outlined to describe the process of exciting electromagnetic fields used for ICR heating of plasma. According to this approach, the electromagnetic fields excited by the induction method in a waveguide (in this case, it is a metallic vacuum chamber) are presented as a superposition of TE- and TM-modes. Based on this approach, a model was later developed of the excitation of electromagnetic fields in a plasma in an external magnetic field and the gain was found for a radial electric field component created by screw antennas [62]. The gain is approximately given by the formula $K \approx 1/(k_z r_A)^2$, where r_A is the radius of the screw antenna. Attention was paid to this effect in patent application [6]. The effect was calculated for the conditions of the French facility ERIC [30]. It was measured in the lithium plasma of the Siren' facility: $K = 5-6$ [63].

The possibility of expanding the plasma column in an ICR installation, which is related to the choice of induction antenna, was mentioned in Ref. [64]. This effect was used in controlled thermonuclear fusion for varying the cross section of the plasma column in toroidal discharges [65]. It arises due to a rotating magnetic field. The effect is not connected with ion cyclotron resonance and can be realized in a wide frequency range, including ω_{ci}. In an ICR installation, it would probably be obstructive because the field rotating in the direction of ion cyclotron rotation causes expansion of the plasma column (the column contracts as the field rotates in the reverse direction). In Ref. [66], a similar effect was observed; however, its influence on the separation process has not been studied. If the influence is substantially obstructive, then a multiphase antenna should be replaced with a single-phase antenna, for example, with the one employed in Ref. [37] (Nagoya Type III antenna with rotation; see Fig. 2).

Finally, we consider collectors for enriched materials. Drawbacks of a traditional collector system (TCS) [6] have already been considered in Section 4. The most important idea concerning TCSs is that of Karchevskii et al. [67, 68], who suggested placing the TCS in the zone of a weakened magnetic field of a facility. In the process of partial transformation of the transverse energy of heated particles into their longitudinal energy ($W_\perp/B = \text{const}$), the deposit expands to a longer distance, i.e., a longer exposure of the collector system becomes possible. In addition, spatial separation of heated and cold ions can be performed because the lead of the screw trajectory of cold ions in a weakened field is shorter than that of heated ions. Initially, no principal changes were stipulated: only the shape and dimensions of the TCS were assumed to change according to the expansion of plasma flux in a reducing field ($BS = \text{const}$). Nevertheless, a collector system specially designed for operation in an axially

symmetric nonuniform magnetic field was later suggested. Radially placed collector plates were used for collecting a product [50]. There are also possibilities to improve the TCS itself in order to increase the separation and extraction factors and make a longer exposure of TCS to plasma [69] (Gorshunov et al.).

A nontraditional collector system based on ion drift in a nonuniform magnetic field has been suggested in patent [70] (Belavin, Zvonkov, and Timofeev). Actually, it is convenient to employ a section of a toroidal magnetic field in which $v_{\rm dr} = c(2W_{||} + W_{\perp})/(eBR)$ [71], where $W_{||}$ and $W_{\perp}$ are the longitudinal and transverse ion energies, respectively, and R is the greater radius of the torus. Thus, in the time needed for passing through the section it is possible to select heated and cold ions that leave the zone of selective cyclotron heating with a plasma flux. In the first approximation, the quantity $W_{||}$ does not change in cyclotron heating. Such a collector system has not been tested with a two-temperature (by the ion component) plasma. It was planned to employ such a collector system in the project [55]. It is also stipulated in the project for reprocessing the waste fuel of fast neutron reactors by the plasma ICR method [72].

6. Conclusions

The main conclusions concerning the essence of isotope separation by the ICR method, which were made in Sections 2–5, entail the practical corollary that a separation ICR facility may become universal if: (a) the ion separation occurs in a magnetic field with an induction of 4–6 T, and (b) there exists a certain way to increase the ion temperature to several electron-volts in ionized metal vapor prior to the onset of the separation process.

A question arises as to why this method is not widely used even though its applicability to isotope separation for all chemical elements with $\Delta M \geqslant 2$ is beyond question. The results of investigations carried out confirm its potential possibilities, especially as investigators have not faced any instability of the plasma produced at the facilities.

The main reason for rare employment of the method is the necessity of using a large superconducting solenoid which is the most expensive part of the setup. In most countries, to create an ICR facility, such a solenoid must be ordered from a specialized company.

In the ISTC (MNTTs) project No. 830 [12], the superconducting solenoid was to be produced in a pilot factory of a large scientific-research institute. It is probable that later on the required solenoid may be created by making a winding from high-temperature superconductors cooled by liquid nitrogen exclusively.

A solenoid with copper windings can be employed in the enrichment of elements with middle mass, for example, calcium. Nevertheless, a vacuum chamber with a diameter of greater than 0.5 m and a zone of the uniform magnetic field about 2 m long are needed for quality enrichment. The solenoid should have the corresponding size; hence, the power supplied to it would be about 1 MW.

It is important that the ICR method will not, generally, compete with the industrial electromagnetic method. The limiting separation factor for the ICR method is $\alpha = 50-100$. Hence, for obtaining pure isotope materials the product obtained should be additionally enriched at electromagnetic separation facilities.

It seems likely that the ICR method will not compete as well with the laser AVLIS method in isotope separation of the chemical elements whose isotopic shift at the first stage of atomic excitation is small compared to the width of the spectral line (elements from potassium to zirconium). The methods capable of performing isotopically selective photoionization of atoms with comparatively high productivity of the facility are not simple in this case; the author of Ref. [73] even calls them 'sophisticated'. Experimental investigations of selective photoionization of calcium (photoionization scheme of 272.1 nm + 678.3 nm) performed at IMP RRC 'Kurchatov Institute' [74] allowed the authors to recommend this scheme for obtaining weight quantities of calcium isotopes. However, difficulties will arise connected with the general background of neutral particles inside the facility [75]. Allowing for this fact, employment of the ICR method seems more acceptable in separating, for example, the rare ^{48}Ca isotope.

Comparison of the details of methods for isotope separation cannot substantiate preference of one method over another. It is necessary to take into account the availability of the corresponding devices, operating costs, etc. It is important to concentrate on the main goal. In order to satisfy present and future isotope demands for those elements that cannot be obtained by the centrifugal method (because they have no chemical compounds possessing high vapor pressure at room temperature), it is reasonable to use either plasma or laser separation methods, depending on the particular task, rather than to challenge them.

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