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ISOTOPE SEPARATION IN PLASMA BY ION-CYCLOTRON RESONANCE METHOD A.E.Dubinov,* I.Yu.Kornilova, V.D.Selemir

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ISOTOPE SEPARATION IN PLASMA BY ION-CYCLOTRON RESONANCE METHOD A.E.Dubinov,* I.Yu.Kornilova, V.D.Selemir

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Contemporary state of investigations on isotope separation in plasma using selective ioncyclotron resonance (ICR) heating is considered. The main attention is paid to necessary conditions of heating selectivity, plasma creation methods in isotope ICR-separation facilities, selection of antenna systems for heating, and principles of more-heated component selection. Experimental results obtained at different isotope mixtures separation are presented.

Рассматривается современное состояние исследований по разделению изотопов в плазме с помощью селективного нагрева в режиме ионно-циклотронного резонанса (ИЦР). Особое внимание уделяется необходимым условиям селективности нагрева, методам создания плазмы в установках ИЦР-разделения изотопов, выбору антенных систем для нагрева и принципам отбора более нагретой компоненты. Приведены экспериментальные результаты, полученные при разделении различных смесей изотопов.

Isotope separation is a large branch of industry. Isotopes have wide application in radiomedicine, agriculture, in atomic power industry and are also used to solve the problems of fundamental physics. The existing industrial isotope separation methods (gas centrifuges, electromagnetic separations, etc.) cannot always suit the requirements of the economy. This is related either with very low efficiency of electromagnetic separation method or with the absence of appropriate volatile matters, necessary for centrifugal separation.

The search for alternative technologies, allowing one to widen the range and assortment of isotope set-ups, being manufactured, and to decrease the power consumption, has led to development of absolutely new isotope selection methods. Among them there are: laser method [1], photochemical method of mercury isotope separation [2], and also plasma separation methods. If the first two methods are studied well enough at present and are aimed at successful application in industrial scale, then plasma methods need further investigation and development. At inevitably relatively high costs of evaporation in plasma facilities, ionization (of the order of 300–3000 eV/atom) of matter and technical difficulties for pri-

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mary effect multiplication, the facility acceptable for scaled production will be considered — the one that has high enough separation factors.

Among the known technologies of separation in plasma, the most prospective for the given tasks decision is the isotope separation method based on isotopic selective ion-cyclotron resonance (ICR) heating of plasma. The detailed enough description of development history, of physical basis of the method and of installations for its realization can be found in the report of 1991 [3]. It should be noted that for the time period after the report publication a large amount of new information was stored. Numerous calculation-theoretical works on the given topic were executed. Besides this, new designs are proposed, which, possibly, will allow hundreds of times increase of the ICR-method effectiveness. The present review is aimed at generalization of the new works with short summary of old material.

In 1975, G.A.Askaryan et al. proposed to use ICR for separation in plasma [4]. Probably, the works in the USA, have been executed: in the late 1976, the results of the first successful experiments on ICR-separation of potassium isotopes carried out at the firm TRW were published [5]. In the USA, the further development of the method was directed at uranium isotope separation. However after the ICR-method was not accepted as the current technology of uranium enrichment in 1982 on economic considerations, that was shortly announced in [6], stable isotope separation has become the basic field of application. After the work [5] was published, for seven years no additional scientific information about ICR-method appeared in the press. The new data were obtained only in 1983 after the reports of American scientists at the two conferences [7, 8]. These works witnessed significant progress in ICR-method development for seven years mentioned. Successful experiments on nickel, indium and lead isotope separation were performed.

The following works were carried out in France [9] on calcium isotope separation and in the USSR on lithium isotope separation. At present the experimental material about ICR-method of separation is practically limited by the above-mentioned publications and several reports at the conferences [7,8,12–14].

Analyzing the method development, the surprising thing is that it has been started so late. The necessary data on ICR were obtained already by the early 60's [15]. Development of ion-cyclotron resonance spectroscopy [16] has started at about the same time. It remains to suppose that the reason for the ICR-method development delay was the imagination of plasma as an extremely unstable substance in which it is impossible to observe fine effects. Nowadays, on the basis of the existing experience on isotope separation with the help of ICR, it is possible to state that magnetized and comparatively cold plasma formed as a result of metal vapors ionization is rather stable. Hence, some imaginations of heating and ion movement on the basis of single particle model are close to reality. Resting on the ICR-method of separation groundwork is presented below.

The principal idea of ICR-method is the following. A flux of collision free plasma comes into heating area in which the aimed isotope ions advantageous acceleration is executed on resonance frequency:

$$\omega = \omega_{ci} = \frac{eB}{M_i}.$$

Here ω_{ci} is Larmor frequency of aimed isotope ions in magnetic field B, M_i is the mass of separated isotope ion. As a result the increase of their transverse energy takes place. Accordingly their Larmor radius $r_{\rm L}$ increases. Selective heating of the desired ion fraction takes place. Isotopes, neighbouring to the yielded one, because of small difference $(\Delta\omega)$ of their own frequency and external effect frequency, meet on their way several units of beats. This phenomenon does not allow them to continuously increase their energy, so their maximum energy will not exceed the following fraction of resonance ion maximum energy: $(T/2\tau)^2$, where $T = 2\pi/\Delta\omega$ is a beats period, τ is the time of flight through heating zone. Thus, at the output of heating zone, plasma ions will be divided to cold and more heated. Because of the difference of their Larmor radii only heated ions will be effectively trapped by the selector (the system of plates, parallel to the flux and magnetic field direction).

To provide heat selectivity it is necessary to realize several conditions. It is obvious the requirement of stable magnetic field uniformity in the heating area:

$$\frac{\Delta B}{B} < \frac{\Delta M_i}{M_i}.$$
(1)

Here, $\Delta B/B$ is a relative change of magnetic field in ICR heating area, $\Delta M_i/M_i$ is a relative resolution to the mass (ΔM_i is the difference of selected and neighbouring isotope masses). To select most isotopes, the value $\Delta B/B$ shall be not more than 10^{-2} . For obtaining good resolution of ICR lines and dealing with rare-earth metal isotopes, the much more uniform field ($\Delta B/B \sim 10^{-3}$) is required. The magnetic field value is selected according to the mass of isotopes being separated with the account of heat velocity. Usually its value does not exceed 3 T.

The other condition of heating selectivity is the requirement of smallness of cyclotron absorption line widening due to the heated ions collisions:

$$\frac{\nu_{ii}}{\omega} < \frac{\Delta M_i}{M_i}.$$
(2)

Besides, the connection of ion–ion collisions frequency ν_{ii} with plasma parameters is defined by the following relation: $\nu_{ii} = 5 \cdot 10^{-7} n_i / \Theta_i^{3/2} \sqrt{A}$, where n_i is heated ions density in cm⁻³, Θ_i is their temperature in eV, A is the isotope mass in atomic units. Usually as a result of estimations to formula (2) the obvious requirement is obtained: during the flight through the heating area a particle shall be subjected to no more than one collision.

Definite requirements are imposed on the ion starting temperature Θ_i . It is reasonable to have low Θ_i , that allows one to achieve larger difference of selected isotope energies and other ion components, and, hence, to realize larger values of enrichment. However, a very low initial energy ($\Theta_i < 1 \text{ eV}$) leads to the increase of the Coulomb collisions frequency $\nu_{ii} \sim 1/\Theta_i^{3/2}$ at the initial stage of heating and, accordingly, to the limits imposed to plasma density in the beam. Accounting to the above-mentioned factors, the most optimal initial ion temperature is considered to be $\Theta_i \sim 2 \div 15 \text{ eV}$.

The next heating selectivity condition is determined by the inequality:

$$\frac{\Delta\omega_{\tau}}{\omega} = \frac{\pi V_z}{L\omega} < \frac{\Delta M_i}{M_i}.$$
(3)

Here $\Delta \omega_{\tau}$ is flight-time broadening of cyclotron absorption line, V_z is an average longitudinal ion speed, L is the heating zone length. When realizing this condition, significant difference between the energies of selected isotopes and neighbouring to it is provided at the output of the heating zone.

Nonzero initial plasma temperature means some spread of the ion speeds in plasma. Having the spread of the ion longitudinal speeds ΔV_z , the Doppler effect influences the heating selectivity:

$$\frac{\Delta\omega_{\rm D}}{\omega} = \frac{\kappa\Delta V_z}{\omega} = \frac{2\pi\Delta V_z}{\lambda\omega} < \frac{\Delta M_i}{M_i}.$$
(4)

Here $\Delta \omega_{\rm D}$ is Doppler broadening, κ is the wave number, λ is the cyclotron wavelength in plasma.

For effectiveness of separation process it is important not only realization of inequalities (3) and (4), but definite relation between passflight $\Delta \omega_{\tau}$ and Doppler $\Delta \omega_{\rm D}$ broadenings:

$$\Delta\omega_{\tau} \geqslant \Delta\omega_{\rm D}.\tag{5}$$

At realization of this condition, the major part of ions is involved into acceleration process. As the fraction of heated ions will be deliberately more than one and not all ions will be collected by the selector, it is real to obtain the ratio of the matter application.

At ion speeds spread in the beam $\Delta V_z \approx V_z$, specific for low temperature sources, it comes out from condition (5) that the most optimal for ion cyclotron heating are antennas with wavelength $\lambda \ge L/2$. The obtained as a result of calculations [17] dependences of resonance curve on the wave number $\kappa = 2\pi/\lambda$ and the heating zone length L, confirm the fact of resonance curve broadening at $\lambda \ge L/2$. The application of antennas with smaller wavelengths degrade heating selectivity also because of nonlinear effects manifestation [18]. Calculations accomplished in this work have shown that simultaneous heating of resonance particles takes place only on the length of the order of average axial transition of particles from plasma source during half period. Then, because of movement phases difference, the average energy of particles tends with oscillations to stable value, and the spectrum of energies becomes maximally wide. Nevertheless, at some conditions [19] the selective heating is also possible in case of nonlinear oscillations captured by HF-field of resonance ions.

Two types of waves being able to carry out isotopically ion selective heating are in use at present. The first type is the so-called «ordinary» electromagnetic wave, the electric field vector of which rotates in the same direction as the ions during Larmor rotation. The other type of waves being able to carry out ion selective heating at a frequency close to ion-cyclotron one, are electrostatic ion-cyclotron waves [20] which have $\kappa \parallel \mathbf{E}$. It is possible to excite them only with contact process, e.g., supplying alternate potential to metal filament stretched along magnetic field to plasma column [21]. Contact process of excitation is unsuitable for industrial applications for some reasons which will be noted. Thus, the other method of electrostatic ion-cyclotron waves excitation is used in experiments most frequently, when induced excited electromagnetic waves are transformed into electrostatic ones [22].

On the basis of conditions (1)–(5), plasma parameters (n, Θ_i, V_z) and installation parameters (B, L, λ) are chosen. It is obvious that limitations imposed on



Fig. 1. Block diagram of the plant for ICRmethod isotope separation: 1 — substance feeding system, 2 — vacuum chamber, 3 — plasma source, 4 — plasma source magnetic coil, 5 — the plant magnetic coil, 6 — HF-antenna, 7 — plasma flux, 8 substance samples selector

It is obvious that limitations imposed on n and V_z are the limitations for the installation output $G = \xi c_0 M_i n V_z S$, where ξ is the substance use ratio, c_0 is the initial selected isotope concentration, S is the plasma flux cross section. Based on acceptable values determining the output of installation for isotope separation, the calculated annual isotope production in a large plant is not less than 500 kg.

Let's step to description of the plants with the help of which isotope separation with ICR-method is executed. As has been noted, there exist only three plants for ICR-method isotope separation [5,9,10], which are designed and built by three experimental groups in the USA, France and Russia. The common ele-

ments of these plants are: 1) vacuum chamber; 2) plasma source producing the flux of ionized atoms of the element the isotopes of which are being separated; 3) the uniform magnetic field; 4) HF-antenna with the help of which in the

uniform magnetic field the isotopic selective ion heating is accomplished at the frequencies close to ion-cyclotron ones; 5) collector for extraction of selectively heated isotopic ions out of plasma flux (see Fig. 1).

The ideas of the experiment execution are like each other, however there are some differences between plasma creation processes, ways of selective ion heating, ways of aimed isotope collection, the rest matter collection, etc.

By present, three methods of plasma creation are used in ICR-plants: ionization of evaporated atoms on the hot surface of refractory metal [5], microwave ionization at the frequency of electron cyclotron resonance [9] and also permanent current discharge in the substance isotope vapors [10]. The first process has significant limitations on plasma density. In most plants the density of the created in this way plasma does not exceed 10^{11} cm⁻³. Some doubts in correctness of such a point of view are induced by the work [23] in which uranium plasma with density higher than 10^{12} cm⁻³ was obtained. The authors of this work tended to obtain more dense, 10^{14} cm⁻³, plasma but the information confirming the forecast did not follow. The disadvantage of discharge-gas source is the comparability of plasma column cross section with cathode cross section, while the design of the latter is accounted to the use of cored cathode. Construction of industrial plants of cored cathode of a large diameter (~ 50 cm) seems to be hardly realized. For large separation ICR-plants, atom microwave ionization is considered to be the most prospective out of the mentioned processes of plasma creation.

As for the ways of ion-cyclotron heating, there are two of them by today: inductive and electrostatic. The application of electrostatic process in which potential difference in plasma is created at the expense of contact with faced electrodes, connected to AC source, is commonly supposed to be no prospect. It is firstly connected with the action of electrostatic mechanism of the field formation only at small concentration of resonance particles in binary ion mixture [24] and also with unsuitable coincidence of antenna and selector functions in one design. It can be stated today, that the inductive method, which is realized by superposition of weak alternate field on permanent uniformal magnetic field, is favoured.

The helical antenna proposed by Hipp et al. [25] is considered to be the most suitable for ion-cyclotron heating; such antenna is the wiring producing rotary electrical field with mode m = 1. Helical antenna has a number of advantages in comparison with solenoidal one. At first, it generates practically uniformal electric field in close-to-axis installation zone that provides uniform heating of plasma jet. Secondly, it produces favourable conditions for volume charge compensation with electron currents along magnetic field.

Solenoidal antenna, which is considered to be unpromising, could be effectively used in separation of some isotopes. This is mentioned in the work [26]. Calculations accomplished in the given work have shown that in spite of solenoid antenna field shielding in plasma, the inductive heating, nevertheless, is enough for separation of isotopes with mass number about 150. However the plants with plasma heating of this kind do not provide the effectiveness which is enough for industrial scales. The calculated in [26] annual effectiveness of the plant for gadolinium isotope separation was about 100 kg of ¹⁵⁷Gd isotope. In calculations the following plasma and separation system parameters were used: heating zone length L = 2 m, plasma jet initial temperature $\theta = 10$ eV, radius a = 10 cm, plasma density in the jet $n = 10^{12}$ cm⁻³, external magnetic field value B = 3 T. Solenoidal antenna excites the HF-field with azimuth number m = 0. Current amplitude in antenna turn $I_0 = 60$ A and full number of turns N = 150 were chosen to provide the obtaining of the ion energy necessary for isotope separation W = 200-300 eV. The calculated voltage on the antenna was 70 kV.

In the experiments, the ion heating was tracked with the help of electrostatical analyzers inserted inside of the plasma column. In large separation plants, the analyzers, possibly, will serve for technological control. For plasma diagnostics in the magnetic field two types of analyzers are used. The first type are multigrid analyzers. Their detailed description can be found in [27, 28]. The analysis of this type analyzer operation applicable to the problem of isotopes separation in plasma is accomplished in the recent work [29]. This type of analyzers allows one to measure the following basic parameters of magnified ions in plasma flux: longitudinal axis of ions, cross-sectional energy spectrum of ions, spatial distribution of ion density in the flux, full current of ions. The other type are more simple by their construction analyzers [30] having only the case and collector, without grids. Inserted into plasma across magnetic field they allow one to carry out ion analysis to cross-sectional energies. The given type of analyzer is the prototype of the selector of isotopically selectively heated particles, used in experiments on isotope separation with the ICR-method.



Fig. 2. Elemental cell of product collector: 1 - front screens, 2 - plates for product collection, 3 - plates for atom capture in case of spent material dispersion, 4 - receiver of depleted plasma flux

The product selector is a system of plates (collectors) located parallel to magnetic field and longitudinal flux of plasma, forming the grid and placed at a distance of the order of 1–1.5 Larmor radii of heated ions to each other. As a result the collector effectively captures the heated ions while the major part of cold ions, Larmor radii of which are less than the dis-

tance between the plates, pass through the collector. A part of cold ions moving at a distance less than their Larmor radius are also captured by selector. The ions which don't get to the plates are collected by the receiver of depleted plasma flux located behind the plates. The elemental cell of collector system is shown in Fig. 2.

Collection of enriched with aimed isotope substance is conducted by evaporation of ions collected on the plates. The rate of distribution achieved as a result of the experiment is determined by the following formula:

$$q = \frac{c(1-c_0)}{c_0(1-c)}.$$

Here c and c_0 are concentrations of extracted isotope in the product and in the initial mixture, respectively. The value q, obtained at zero confining potential on the plate, is called «geometrical» separation factor.

In order to reduce the flux of cold ions to aimed isotope collector, it is supposed to supply repulsion potential to the plates. Besides this, in front of the plate face, crossly located screens can be installed which do not let ions with Larmor radius less than the screen's height through. The choice of the screen's height needs optimization, as significant increase of their height allows significant decrease of the limited value of extraction of the isotope being enriched $\gamma_{\text{max}} = (b - 2a)/b$, where a is half height of the screen, b is the distance between collector plates. On the other hand, small screen height, or its absence, leads not only to increase of the cold ions flux to selector, but also to dusting the plates with neutral particles formed during recombination. The following system parameters are also to be optimized: collector plate length z, the distance between them b, the value of positive repulsion potential U_k .

Dependences of the ion flux to collector on concentration of the substance dispersed on the plates at the given parameters were investigated in several works. In [31] a method of calculation of the ion flux to collector system was proposed. It consists in introducing the distribution function of accelerated particles to crossing velocities with the following calculation of fluxes to collector by means of numerical integration along longitudinal (V_z) and cross-sectional (V_{xy}) to magnetic field velocities and also to cross-sectional coordinate of the particle leading centre y_0 . The function of distribution function is equilibrium. The function of distribution to cross-sectional velocities was approximately calculated on the basis of the Boltzmann equation for collision free plasma in supposition that the initial distribution function is equilibrium. The function of distributions were performed applicably to separation of binary mixture of lithium isotopes by single-wave antenna. The obtained dependences of Li flux density, ⁶Li extraction factor and ⁶Li concentrations on parameters of collector system have shown, on the whole, a qualitative coincidence with the investigation results.

In [32] calculations of flux density and concentration of the aimed isotope dispersed on the plate were carried out accountless to the effect of the repulsion potential on the collector. In this work the modified calculation methods were used containing in primary definition of ion effective temperatures with ensuing

calculation of ion fluxes to collector. Unlike to [31], the distribution functions of heated and cold ion to cross-sectional energies were supposed to be Maxwellian with different effective temperatures. The function of distribution to longitudinal velocities was selected the same as in [31], equilibrium, with the absence of particles moving towards ion resource. The full distribution function of particles of any sort at the given suppositions is the following:

$$F = 2n\left(\frac{M_i}{2\pi kT_{xy}}\right)\left(\frac{M_i}{2\pi kT_z}\right)^{1/2} \exp\left(-\frac{M_i}{2k}\left(\frac{V_{xy}^2}{T_{xy}} + \frac{V_z^2}{T_z}\right)\right), \quad V_z > 0.$$

Here T_{xy} and T_z are cross-sectional and longitudinal ion effective temperatures in plasma, k is the Boltzmann constant. For calculation of the ion distribution function applicable to binary isotope mixture ⁶Li-⁷Li, the authors used the results on the obtained calcium isotope mixture [33]. The installation parameters used in calculations were taken from [10]: B = 0.25 T, $E_0 = 54$ V/m, L = 0.8 m, $T_{xy} = 5$ eV, $T_z = 10$ eV.

In the work [34], being the continuation of [32], the effect of positive repulsion potential on the collector has been used during selection of heated ions from plasma flux. Dependences obtained in [32] and [34] reflect the same qualitative ideas that in [31]. In Figs. 3–6 specified dependences are presented, which allow one to make the following basic conclusions.

The simple difference between Larmor radii of resonance and nonresonance ions, connected with difference of ion effective temperatures for real modes of isotopical-selective ICR heating, leads to separative effect during the substance deposition on the collector. However the value of this effect is usually insufficient for practical tasks. The use of the front screens and positive repulsing cold-ions potential on the collector allows one to increase significantly the concentration of the aimed isotope, being dispersed on the plates.

The condition of ion pass-flight through the front screen is the following:

$$a < y_0(1 - \cos \alpha) + \sin \alpha \sqrt{r_{\rm L}^2 - y_0^2} < b - a.$$
 (6)

Here $a = \omega z_1/V_z$ is the angle of the ion turn during the pass-flight from the plane of the screen to the place of collision with the plate, z_1 is longitudinal coordinate of the place of ion collision on the plate surface, ω is the frequency of ion cyclotron rotation, r_L is ion Larmor radius. This condition is realized at large enough Larmor radius $r_L > a/(2 \sin \alpha/2)$ in limited coordinate range of leading centre Δy_0 [32]. Thus even heated enough ions, the leading centre coordinates of which do not satisfy the condition (6), get to the screens with following dissipation or condensation. Hence the reduction of the flux density of the aimed isotope to the plates takes place. Separation system parameters allow one to evaluate the optimal height of the screen at which the ratio between concentration increase effect and density decrease effect is more favourable. In [32], for comparison, the results obtained at a = 0 and a = 3 mm are presented. It is necessary to note that at a = 3 mm the decrease of the flux density is insignificant (~ 1.25 times) in comparison with the concentration increase (~ 1.6 times) (see Fig. 3). In this case the height of the front screen practically coincides with the value of Larmor radius of nonresonance ions (~ 3.6 mm). The further screen height increase leads to equating the effects of concentration increase and flux density decrease, so it is reasonable to use the front screens with the height of the order or less than cold ion Larmor radius.



Fig. 3. Dependences of dimensionless flux density of ion ${}^{6}\text{Li}^{+}(a)$ and isotope ${}^{6}\text{Li}$ concentration (b) on longitudinal collector coordinate for different heights of the front screen: l - a = 0; 2 - a = 1.5 mm; 3 - a = 3.0 mm ($b = \infty$, $U_k = 0$) [32]

Positive repulsing nonresonance ions potential U_k applied to collector plate helps more uniform extraction of resonance component from plasma flux. The ions having significant velocity component normal to collector surface, overcome potential barrier and condense on the plate. Other ions are reflected and get out of the flux. Condensation condition can be written in the form:

$$M_i(V_u^2/2) \geqslant eU_k. \tag{7}$$

Here V_y is the ion velocity in the plane perpendicular to the plate. From condition (7) we obtain the following limitation on the range of leading centres coordinates:

$$-\sqrt{r_{\rm L}^2 - \frac{2eU_k}{M_i\omega}} < y_0 < \sqrt{r_{\rm L}^2 - \frac{2eU_k}{M_i\omega}}.$$
(8)

Thus, part of ions having significant energy but flying up to collector along soft trajectories will get out of condensing flux. This phenomenon again helps the reduction of aimed ion flux density to collector. However, this effect simultaneously significantly increases the concentration level of isotope dispersed to the plates. For example, the presented in [34] results show that at $U_k = 40$ V it is possible to achieved nearly 100% enrichment with isotope ⁶Li (for comparison at $U_k = 0$ ⁶Li concentration value is 20%); ⁶Li⁺ ion flux density to collector decreases only 2.5 times (see Fig. 4). It should be noted that there is no sense using maximum allowable value U_k , as significant part of cold ions with crosssectional energy W_y is already cut off at U_k a little larger than W_y . Further increase of collector potential will help not only cutting off nonresonance ions, but decreasing the aimed isotope ions flux to the plate. So it is undesirable to select the value U_k one–two tens more than the value of cross-sectional cold ion energy.



Fig. 4. Dependences of dimensionless flux density of ion ${}^{6}\text{Li}^{+}(a)$ and isotope ${}^{6}\text{Li}$ concentration (b) on longitudinal collector coordinate for repulsing potentials U_k : a) $I - U_k = 0$; $2 - U_k = 10$ V; $3 - U_k = 20$ V; $4 - U_k = 40$ V; b) $I - U_k = 0$; $2 - U_k = 10$ V; $3 - U_k = 20$ V; $4 - U_k = 30$ V; $5 - U_k = 40$ V [34]

The next collector parameter, affecting the rate of the substance distribution, is the distance between collector plates b. As was noted above, the most optimal value b is in the range $1-1.5r_L \div 1.5 r_L$, where r_L is Larmor radius of heated ions. Notable reduction of the distance b in comparison with r_L leads to changing the sign of mixture enrichment to collector length z. This phenomenon was observed in [32] in case of enrichment with isotope ⁶Li already at b = 7.5 mm while the Larmor distance of ⁶Li ions was equal to 15 mm (see Fig. 5). Significant increase of the distance between the plates (from $2-3 r_L$) was accompanied by significant reduction of the flux density of the extracted isotope at small increase of concentration, which is not reasonable and so is extremely undesirable.



Fig. 5. Dependences of dimensionless flux density of ion ${}^{6}\text{Li}^{+}(a)$ and isotope ${}^{6}\text{Li}$ concentration (b) on longitudinal collector coordinate for different distances between collector plates b: I - b = 30 mm; 2 - b = 15 mm; 3 - b = 7.5 mm [32]. Uniform line (4) is initial ${}^{6}\text{Li}$ concentration ($a = 0, U_k = 0$)

While choosing collector plates length it is necessary to take into account the following facts. The heated ions flying to the beginning of collector plates have different coordinates therefore until the moment of collision with the plate they are to necessarily turn at different angles $\alpha_i = \omega t_i$, where t_i is the ion time-of-flight distance from the screen plane to the place of collision. Maximum angle of turning is defined by the value 2π . Thus, the collector plate length, z, is to be chosen such that during the flight along the plate the ions could have time to make full turn-round along Larmor helix:

$$z = V_z t = 2\pi z = \frac{V_z}{\omega}.$$
(9)

Besides, all resonance ions, the leading centre of which is at a distance $y \leq r_{\rm L}$ from collector, will get to the plate. In case of enrichment with ⁶Li isotope at characteristic longitudinal velocity $V_z \approx 10^4$ m/s, the value of z will be ~ 15 mm. Special attention needs the fact that maximum concentration of the isotope, being enriched by more than 80%, is obtained at once behind the screen and reduces practically to the zero value at the end of collector plate. Therefore to increase average concentration of the isotope, being dispersed, one needs to choose the optimal collector length less than the one calculated by the formula (9). The reduction of collector length shall be performed with the account of dependence of target isotope extraction factor on z (see Fig. 6). It is necessary to take into account that significant reduction of z leads to significant reduction of the matter extraction factor.



Fig. 6. Dependences of ⁶Li flux density (1), ⁶Li extraction factor (3) and ⁶Li concentration (2) on longitudinal collector coordinate z [31]

The above-mentioned analysis of the sampler parameters of the separation system does not leave any doubt that only geometric factor is manifestly insufficient to obtain highly enriched mixture. Therefore, as a rule, in all plants on isotope separation by ICR-method, positive repulsive potential is used applying to collector plates in combination with the front edge screens. However, introduction of the screens and detaining potentials is not the optimal decision to increase isotope separation ratio. Their use though allows one to increase separation ratio several times but simultaneously leads to reduction of operation matter usage factor. Besides, this positive detaining potential can lead to collector plates heating and

to evaporation of the collected isotope. To eliminate the enumerated problems the qualitatively new approach to the given task is needed.



Fig. 7. Block diagram of the principal operation of the plant on ICR-method of isotope separation with the portion of nonuniform magnetic field: L_1 — the length of the nonuniform magnetic field portion, L — heating zone length, I — plasma flux, 2 — the source of electrons, 3 — the flux of selectively heated ions, 4 — target isotope collector, 5 — basic plasma flux, 6 — collector of depleted plasma flux

In [35], it was proposed for the first time the design advance, allowing to increase isotope separation ratio by more than two orders. Technically the given task is realized with the help of the device the principal operation of which is shown in Fig. 7. The difference in the design of the proposed plant is the portion of nonuniform magnetic field placed after the heating zone in which spatial separation of heated plasma ion fraction from the cold one takes place. At this portion the ions displace (drift) in the direction across power lines of the nonuniform magnetic field $V_{dr} = W/eBR$, where W is the energy of perpendicular to magnetic field B ion movement, R is magnetic field power lines

curvature radius. The nonuniform magnetic field parameters are chosen in such a way that the following condition is satisfied:

$$V_{dr}\frac{L_1}{V_z} > d.$$

Here L_1 is the length of the nonuniform magnetic field area, d is plasma flux diameter. While meeting this condition, resonance ions leave the area occupied by plasma, whereas nonresonance displacement ions stay at basic plasma flux (because of their energy small in comparison with the resonance one). Spatial separation of ions of various isotopes allows one to carry out their recombination in far enough and shielded from each other parts of the plant, reducing the possibility of getting one isotope to the collection place instead of the other and by this to essentially increase separation ratio. While creating such a plant, one needs to take into account plasma ions spatial separation, causing the ions ejection to the chamber walls in the crossed electric and magnetic fields. Therefore additionally there should be created a compensating electron flux along magnetic field power lines for positive charge neutralization.

The authors [35] have calculated the minimum length of the nonuniform magnetic field at which isolation of target isotope ions from plasma flux takes place. Its value was ~ 0.35 m at the following parameters: $B \approx 0.25$ T, $R \approx 1$ m, $V_{dr} \approx 0.35 \cdot 10^3$ m/s, d = 0.2 m, $V_z \approx 0.6 \cdot 10^3$ m/s.

Disadvantage of the device proposed in [35] is comlicacy of realization with its help of the repeated separation in one vacuum cycle of plasma flux separated at the first step of separation and also the presence of the long portion of nonuniform magnetic field. Technical decisions of the above-described problems are proposed in [36,37]. Application of circular plasma fluxes [36] allows one to reduce the length of nonuniform magnetic field several times, as for full separation of heated and cold ion fractions in the given case it will be enough to displace them relative to each other not for plasma diameter but for thickness of square equivalent ring. Besides this, the use of circular plasma sources helps to increase ICR-heating effectiveness as in this case depth of high-frequency field penetration into plasma reduces and, thus, more uniform heating of plasma flux is obtained.

The proposal to use configuration of nonuniform magnetic field [37] including even number of toroidal solenoid portions with alternating on the curvature sign (see Fig. 8) gives one the possibility to realize in one vacuum cycle multistage separation at the expense of periodical returning of the plasma flux in its original position. The system of movable collector, placed between portions of toroidal solenoids, allows yielding ions of any of the two plasma sources. Not yielded flux in the next toroidal portion with opposed curvature returns to the original position and can enter similar to the previous stage of mass-separation. The use of multistage process of isotopes separation in plasma realized in one vacuum cycle allows one to increase essentially separation ratio and, thus, to obtain more pure matter. Suppositionally, with the help of the proposed design one can reach the enrichment factor up to 10^4 , which more than 100 times increases the similar value, obtained in experiments [5,9,10].



Fig. 8. Schematic image of the device with portions of nonuniform magnetic field for isotope separation with ICR-method in plasma: 1 - vacuum chamber; 2 - plasma source; 3 - electronic emitters; 4 - plasma flux; 5 - solenoid of nonuniform magnetic field; 6 - electron flux; 7,9 - solenoids of nonuniform magnetic field with different curvature sign; 8 - the matter selectors

One more method of increasing isotope separation ratio with ICRmethod was suggested in [38]. This method is based on spatial separation of mixture components and consists in that the use of traveling waves with cyclotron frequencies leads to ions drift of gap isotope both perpendicular to the direction of these waves propagation and to the lines of external magnetic field. Thus, partial spatial isotope separation takes place. It allows one to decrease the possibility of hitting of one isotope into the place of another, i.e., to increase the separation ratio.

To finish the review of ICRmethod of separation we present briefly the results of experimental work in this field. As was noted, the experiments on separation of kalium, calcium and lithium isotopes, accomplished in the USA, France and USSR before 1990, cover practically the whole experimental material. Their detailed discussion and the schemes of separation plants can be found in the review [3] of 1991.

Any isotope separation method is characterized by the obtained separation ratio q. This value together with energy consumption is the basic criterion for the choice of one or another plant to produce the enriched isotope matter on industrial level. Thus, the results of the experiments on isotope separation by ICR-method shall be considered from the point of view of separation ratio obtained.

In the first experimental work [5] maximum ⁴¹K isotopes separation ratio was found: $q \approx 53$. This more than ten times enrichment of ⁴¹K isotope was obtained by heating the ions on cyclotron frequency ⁴¹K at the value of magnetic field B = 0.4 T. In [10] while separating binary isotopic mixture ⁶Li⁻⁷Li, the enrichment of the mixture with ⁶Li isotope up to 88% was reached at its original concentration of 8%. Such concentration value corresponds to separation ratio q = 84. At this point the frequency of the heating field was $\omega = 640$ kHz, magnetic field value was B = 0.25 T. Nowadays, the record value of separation ratio q = 133 was reached by the French group in experiments [9] on separation in calcium isotopic mixture with natural content of isotopes with mass numbers 40, 42, 43, 44, 46, 48. In the experiment, the following plant parameters and plasma properties were used: $E_0 = 62$ V/m, B = 0.2 T, $\theta \approx 2$ eV, L = 1.7 m, $V_z \approx 10^4$ m/s.

It is easy to notice that in all experiments the isotopes of light chemical elements were separated. It is explained by the fact that at the increase of isotope atomic masses the process of their separation becomes more and more complicated because of the difficulties in obtaining necessary mass resolution. In spite of this problem, successful experiments on selective ICR-heating of the ions of heavier isotopes demonstrate real possibility of application of ICR-method of separation to the isotopes with larger atomic mass. For example, in [5] selective heating of ions of Xe isotopes was carried out, in [7, 8] selective excitation of ions of Ni, In, and Pb isotopes was observed. Apart from experimental works, calculations carried out in [26, 39] demonstrate the possibility of application of ICR-method to separation of heavy isotopes, in particular gadolinium.

The ions of other chemical element isotopes were also subjected to isotopic selective heating, e.g., ions of He isotope in helium-krypton plasma or ions H^+ in hydrogen (H^+, H_2^+) plasma [40, 41]. Well resulting ion-cyclotron resonances for some positive ions of Ne and Ar isotopes and for some separate negative ions of Cl isotopes were observed in [5]. Thus, spectrum of chemical elements, to which ICR-separation method is applicable, is very large. Theoretically this method allows one to separate isotopes of elements with any atomic weight. The only limitation is that the element being separated at room temperature is in solid state and is a conductor. Practical difficulties on separation of heavy chemical elements isotopes related to mass resolution can be solved by the choice of corresponding parameters of the separation system.

As is seen, ICR-method of isotope separation is very attractive because of high separation factors in one separation cycle. However, there are some reasons according to which the method is not yet used on industrial level. Firstly, even at high separation factors the ICR-method does not compensate energy expenses, and, thus, is not profitable. The given problem can possibly be decided by creation of new designs of separation plants proposed in [36, 37] which promise to increase isotope separation ratio more than 100 times at simultaneous energy consumption reduction. The other problem consists in the search for suitable plasma source. Possibly preference is already given to microwave ionization of atoms. In this case one needs much efforts to be applied to develop the source design of large scales with specific parameters, exceeding several times ones obtained in [7].

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