

The Status of the Isotope Separation by PSP

Yuri A. Muromkin

Molecular Physics Institute, NRC "Kurchatov Institute", Moscow 123 182, Russia

Received: April 26, 2012 / Accepted: July 12, 2012 / Published: February 28, 2013.

Abstract: Experiments, projects and patents, concerning practical usage of a separation method, based on ICR (ion cyclotron resonance) in plasma, carried out in the 2000s in USA, EU and in Russia, are discussed in this article. It was planned to use ICR method for production of isotopically enriched burnable poisons for nuclear reactor fuel, and also to use this method for extraction of neutron-excess isotopes for the purpose of observation of neutrinoless double beta decay ($\beta\beta$ 0v). One more application of this method can be a nuclear waste treatment technology. Current situation with realization of these plans is considered.

Key words: ICR (ion cyclotron resonance), selective ICR heating of ions in plasma, isotope separation.

1. Introduction

The first experimental work, devoted to isotope separation, based on ICR (ion cyclotron resonance) in plasma, had been published at the end of 1976 (Dawson et al. [1]). It had been preceded as by experimental investigation of Bernstein waves dispersion in potassium plasma [2], when separate cyclotron damping of these waves were observed at cyclotron frequencies of two ions—potassium isotopes (³⁹K⁺, ⁴¹K⁺), as by proposal of theorists [3], to use cyclotron instability of plasma, confined in a magnetic trap, for mass separation of ions. In the latter work, the principal conditions for selective cyclotron heating of ions with different masses were listed. Numerous investigations in the field of controlled thermonuclear fusion, where cyclotron heating was used, hardly contribute to development of ICR method of isotope separation, because they were made usually on installations with non-uniform magnetic field, and in these works narrow lines of cyclotron resonance were not observed. Thus, the history of isotope separation method based on ion cyclotron resonance comprehends a time period close to 40 years. However, only the first half of this period is

Corresponding author: Yuri A. Muromkin, Ph.D., senior research fellow, research fields: plasma physics and isotope separation. E-mail: ymuromkin@imp.kiae.ru.

characterized by creation almost all of experimental installations. So, already to the beginning of the 1980s in USA, a large separation ICR installation with unique several meters long superconducting solenoid had been created [4]. It was called "PPM (production prototype module)". The ICR separation process itself is called in USA "PSP (plasma separation process)" until now. In the mid 1980s in France (Saclay), investigations started on the installation ERIC (Experience de resonance ionique cyclotron), also equipped with superconducting magnet [5]. In the 1990s, two more installations were used: SIRENA in Russian National Research Center Kurchatov Institute (Moscow) [6]. and in the UCLA (University of California at Los Angeles [7]). In latter, two installations simple "warm" coils were applied for magnetic field creation.

It is necessary to recall a principle of ICR separation method. An ICR separation process scheme was actually presented in Ref. [1]. Its design was formalized in patent by TRW Inc., where PPM was created [8]. Fig. 1 describes principals in experimental installations setup.

Process is started with creation of a plasma flow, in which ionic component is formed by ionized atoms of an element, which isotopes are to be separated. Plasma spread along constant magnetic field B. In the region of

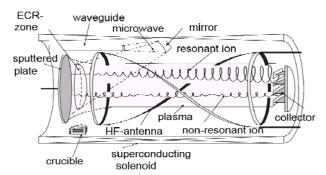


Fig. 1 Scheme of ICR separation process.

uniform magnetic field:

 $(\Delta B/B < \Delta M/M)$, where ΔM —mass difference between isotopes to be separated, M—their mean mass) selective cyclotron heating of the target isotope is performed on the frequency, shifted due to the Doppler effect from $\omega_{ci} = eB/M$: $\omega = \omega_{ci} \pm k_z v_{iz}$. Here, k_z —wave vector, v_{iz} —longitudinal ion velocity (it is assumed, that magnetic field is directed along z axis).

Generally, for creation of electric field, heating ions, i.e., rotating in the direction of Larmor rotation of ions, E_{+} , inductive antennae were used (an antenna, used in ERIC installation is shown in Fig. 1). Their length ranged from one meter to several meters dependent from the mass and mass difference of the isotopes to be separated. Transverse energy of the ion after the time of flight through the heating region $\tau = L/v_{iz}$ is $W_{\perp} = E_{\perp}^2 e^2 \tau^2 / 2M$. Since masses of isotopes are close to each other, the electric field causes periodic increase of energy also of ions-isotopes neighbor to extracted one. The period of beating $T_b = 2\pi/\Delta\omega$, where $\Delta \omega = \omega_{ci} \cdot \Delta M/M$ —difference between cyclotron frequencies of ions of selected and neighbor isotopes. The beating period on the trajectory of a non-resonant ion is $l_b = 2\pi v_{iz} M/(\omega_{ci} \Delta M)$. Therefore, for isotope separation, long enough RF antennae should be used $L > l_b$. Accordingly, condition for selective heating will be: $2\pi v_{iz}/L\omega_{ci} < \Delta M/M$ [1]. Many estimations made with one-particle approximation formulas appears to be correct, because low-temperature magnetized plasma used in ICR separative installations is usually stable (as long as there was only one exception to this rule, according from Chen [9]).

Behind the region of heating, the heated ions are deposited on plates of a collector system, placed parallel to magnetic field strength lines (Fig. 1). In terms, used for separation devices description, this deposit is called a "product". The collector system is served also as an acceptor of the plasma flow, depleted by a target isotope, i.e., "waste". For selection of ions by their energy, positive retarding potential V_r is applied to the plates, and also geometric factor is used: a gap d between the plates, collecting the heated ions, is taken close to double Larmor radius of the heated ions, $d \approx 2r_{Lh}$ ($r_L = v_i \perp /\omega_{ci} = M_i v_i \perp /eB$, where $v_i \perp$ —transverse relative to magnetic field component of ion velocity), and also screens are used with height h, equal to mean Larmor radius of cold ions r_{Lc} .

The key elements of the separation ICR installation are the long solenoid, vacuum chamber and plasma source. The antenna, used for selective heating of ions, and the collector system are less complicated in manufacture.

A quite proper technique for plasma creation in ICR installation is usage of microwave ECR discharge, initiated at electron cyclotron frequency $\omega_{ce} = eB/m_e$, for ionization of atoms [10]. In the case, when sputtering of the target by ions is used for neutral atoms production, a plate, made of a metal, which isotopes are to be separated, is placed in the vacuum chamber at the end of the solenoid.

A negative electric potential |V| = 2-4 kV is maintained at the plate, and microwave radiation with frequency, matching to electron cyclotron resonance conditions at the region several centimeters distant from the surface (ECR-zone), is directed to it. Since the sputtered plate is situated at the end of the solenoid in reduced magnetic field, then electronic component of the produced plasma appears in the trap between the magnetic mirror and negatively charged plate (in other applications the ECR discharge is excited between two magnetic mirrors). A trap is also useful to be created in the case, when neutrals are produced by thermal evaporation (crucible at Fig. 1). For this purpose, a

metal plate should also be placed near the ECR zone, but under negative potential about several tens of volts, that can not cause sputtering.

Plasma escape from the source to the RF heating region starts with escape of electrons to the mirror loss cone, and then turns into an ambipolar process. It is accepted, that plasma travels with ion sonic velocity: $v_z = ((T_e + T_i)/M_i)^{1/2}$.

Generators of microwave radiation, suitable for highly productive installation, i.e., providing 10 A of equivalent ionic current, are gyrotrons or powerful klystrons. However, it will be desirabled to find a method to create plasma flows without using these expensive devices, which have also limited operating life.

All experimental material, important for practical application of ICR separation method, had been obtained at these four referred installations [4-7]. Basic work [1] had been still far from practice. Apparently, it can be explained by commercial interests, that Ref. [4] are the only scientific publication on PPM experiments results. Technical report on the enrichment of nickel by isotope ⁶²Ni [11] is known, but there is no available description of the experiments on enrichment of palladium by isotope ¹⁰²Pd. People only know the amount of enriched palladium obtained in these experiments -40 g [9]. Enriched palladium was used by Theragenics Corporation [12]. Chen [9] makes it clear and certain other details of the experiments in the TRW Inc.. Results of experiments at ERIC and SIRENA installations have been published [13-20]. Photographs of installations are presented in Figs. 2 and 3. Only brief publication exist about experiments in University of California [21].

The works, concerning a research period of ICR isotope separation, are described in detail in Ref. [22].

At the end of the 1990s, a question arouse of creation of modern ICR installation, a new prototype for a highly productive module. Analysis of experiments showed, that in the prototype installation PPM, created at the beginning of the 1980s, magnetic field is insufficient for universal separative ICR installation (magnetic field

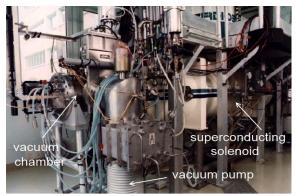


Fig. 2 General view of ERIC installation [5].

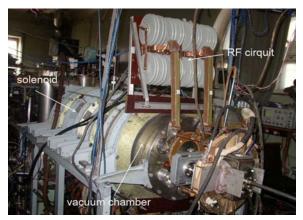


Fig. 3 ICR installation for lithium isotope separation [6].

induction, created by a superconducting solenoid did not exceed 1.8 tesla). Besides, proposals appeared about improvement of separation process itself [23, 24]. Thus, an ISTC project # 0830, 1999-2000 [25] appears.

During the 2000s, other projects have been developed. Characterizing the up-to-date situation, all period of the 2000s should be considered. The next, main part of this article is devoted to this period.

2. Results of the Recent Research and Development Activity in the Field of ICR Separation

2.1 USA

In 2000-th year, it was reported that DOE (U.S. Department of Energy) leased PSP installation to Theragenics Corp. (NYSE:TGX). It was carried out in the frames of DOE Reindustrialization Program. Theragenics is a producer of medical preparation TheraSeed. In this preparation radioactive isotope

¹⁰³Pd is used, which can be produced either by irradiating of monoisotopic rhodium by protons ¹⁰³Rh (p, n), either by irradiating of palladium, enriched by ¹⁰²Pd isotope, by neutrons. It was reported that production of ¹⁰²Pd in amounts, allowing to double TheraSeed production, is one of the aims of PSP leasing. Successful previous experiments on palladium enrichment [9] obviously encouraged the leaseholder. Besides, profitable manufacture of manifold isotopic production was planned [12].

However, only in 2004, Theragenics started palladium enrichment. Until that, the leased installation was used in works on DOE project (NERI Project N 99-0074) "Development of improved burnable poisons for commercial nuclear reactors".

A problem of production of gadolinium, enriched by ¹⁵⁷Gd isotope was tasked continually during the 1990s, not only for ICR method [13], but also for laser AVLIS method [26]. In the last article was called and the reasonable price of enriched gadolinium, \$20/g. As it is known, natural gadolinium is used as a burnable poison in the fuel of nuclear power reactors. Its replacement by gadolinium, enriched by said isotope can give economic profit, and also is expedient for safety reasons.

The project had three phases: selection of elements, suitable for usage as burnable poisons [27], then their isotopic enrichment using PSP device (Fig. 4), and then searching for effective method of their introduction into fuel cladding. According to the results of the work [27], gadolinium, dysprosium and erbium were chosen for isotopic enrichment (by ¹⁵⁷Gd, ¹⁶⁴Dy and ¹⁶⁷Er, accordingly). Calculations show that usage of enriched dysprosium can provide more significant extension of the fuel cycle beside gadolinium. In this paper, by tradition, the author will mainly deals with the gadolinium enrichment.

Results of the experiments were presented in report, prepared by Tennessee University [28]. Before the beginning of the experiments in Ref. [27], preliminary estimations appear of quality and quantity of the expected product (Table 1).

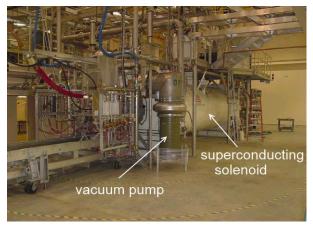


Fig. 4 Layout of American PSP installation [28].

Table 1 Enrichment percentage and annual production for candidate isotopes by plasma separation [27].

Isotope	Natural abundance (%)	Enrichment (% per pass)		Production rate (kg/yr)	
•		2-Tesla	6-Tesla	2-Tesla	6-Tesla
¹⁵⁷ Gd	15.65	45.8	72.5	51	130
¹⁴⁹ Sm	13.8	45.8	71.7	43	109
¹⁶⁷ Er	22.9	55.5	78.9	79	203
¹⁶⁴ Dy	28.2	63.1	83.6	96	245
¹⁷⁷ Hf	18.6	46.0	71. 9	68	175
¹⁸⁴ Os	0.02	0.14	0.41	76	195

Gadolinium of desired quality with $C(^{157}Gd) \ge 70\%$, $\alpha \ge 12$, 5, according to calculations, could be obtained in the field with induction 6-Tesla. In the field of available installation (1.8 T), according to the same calculations, maximum value of separation coefficient for gadolinium isotopes $\alpha \approx 4.5$, and concentration of the isotope in the product $C(^{157}Gd) \approx 45\%$.

The experimental results partially confirmed the prediction. The values of enrichment coefficients, in fact, turned out to be low, even lower than expected and consisted $\alpha = 3.0$ (Gd), 5.3 (Dy) and 2.0 (Er). An amount of enriched matter appeared to be an order less than calculated.

The main reason for the low selectivity of the ICR process in these experiments, the author rightly considered a high frequency of the ion-ion collisions in the plasma v_{ii} . The important condition for the selectivity is in the following:

 $v_{ii}/\omega_{ci} < \Delta M_{i}/M_{i}$ [1, 3]. According to Grossbeck et al. [28], increasing the magnetic field up to 6 T would

provide sufficient selectivity and productivity. Clearly, this requires a new superconducting solenoid, and probably a new installation as a whole, the cost of which exceeds the cost of PSP (Fig. 4).

The reason for the high values of v_{ii} in these experiments can be low temperature of the ions in the plasma and a significant number of doubly charged ions. Collision frequency of single-charged ion with multi-charged $v_{ii} \sim n_i Z^2/T_i^{3/2}$. These reasons were discussed in Refs. [29, 30].

As it was already mentioned, the installation works for palladium enrichment by isotope ¹⁰²Pd in 2004. Production of enriched palladium takes three quarters. In August 2005, exploitation of ICR installation at Theragenics stopped. It was reported that abandoning of PSP is connected with purchasing of CP Medical Corp., of more profitable enterprise. In 2006, the installation was returned to DOE. This information is available in TGX annual reports.

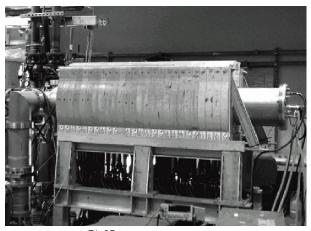
In August 2008, a meeting was held on national requirements in isotopes [31], organized by DOE.

Among the contributions, there was a report by Wong [32], co-author of the work with rubidium enrichment in UCLA [21] and fundamental work [1].

A new ICR installation was presented in the report, with a small superconducting solenoid, created in one of laboratories outside UCLA (Nonlinear Ion Dynamics, LLC), and also a photograph was presented of its prototype (Fig. 5). Apparently, the prototype is the installation used in Ref. [21].

If acquainting with parameters of huge ICR installation, proposed by the author of this report (Table 2), a question arises: how selective heating can be performed at concentration of ions $n_i = 1 \times 10^{14}$ cm⁻³ and at temperature $T_i = 2$ eV? The value v_{ii} is about 1,000 kHz. At such collision frequency selective heating of ions-isotopes at cyclotron frequency f_{ci} about 300 kHz is impossible (point out the misprint in Table 2).

Carty et al. [21] realized in experiments Dawson's proposal [33] to use magnetic mirror for separation of cold and heated ions (heated ions will reflect from the



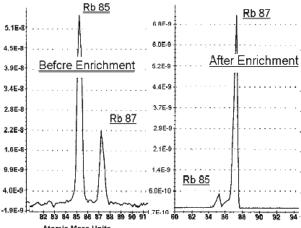


Fig. 5 Installation for separation of rubidium isotopes and experimental results [32].

Table 2 Device parameters [32].

Parameter	Value			
Plasma density	$\sim 1 \times 10^{14} \text{ cm}^{-3}$			
Ion temperature	~2 eV			
Ion drift rate	98,000 cm/sec			
Plasma diameter	1 m			
Particle flux	$7.7 \times 10^{22} \text{ sec}^{-1}$			
Throughput	92 kg/hr			
Device length	~5.5 m			
Magnetic field	4 T			
Electron gyro frequency	90 GHz			
Ion gyro frequency	~30 kHz			
Assuming average mass of 200				

mirror). Probably, this way is unsuitable for a large installation, where transverse dimensions of the plasma column are much larger than Larmor radius of heated particles: for deposition of selectively heated ions, reflected from the magnetic mirror, only a peripheral ring collector can be used. Traditional collector system is placed into the plasma flow, cutting it (Fig. 6).

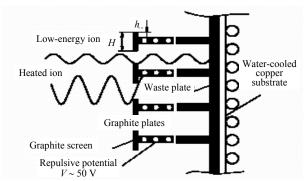


Fig. 6 Traditional collector system [28]. (H and h—outer and inner dimensions of the screens).

Using a magnetic mirror in the selection of heated ions, apparently bound and Wong's project "Mirror-Enhanced Plasma Separation Process (MPSP) for Production of Large Quantities of Isotopes" supported by the DOE in 2009 [34]. Judging by the amount of funding (\$750,000.00), it is a less ambitious project than the example in Table 2.

2.2 European Community

About five years, a question is discussed about application of ICR method for selection of isotopes, perspective for observation of double β decay (48 Ca, 150 Nd). These discussions were held in the frames of ILIAS (integrated large infrastructure for astroparticle science). The double β decay is an unique process for neutrino mass measurement. But this event is very rare ($T_{1/2}(\beta\beta0\nu) > 10^{24}$ years), therefore, it is a question of extraction maximum amount of isotopes: kilograms of 48 Ca isotope, natural abundance 0.185%, tens of kilograms of 150 Nd isotope (5.62%). For achievement of the result, usage of even tons of neutron-excess isotopes should be an advance.

In existing projects (NEMO, GERDA and others), isotopes are used, obtained by centrifugal method. Isotopes of alkali-earth and rare-earth elements can not be extracted by centrifugal method, because they have no chemical compounds with sufficient enough vapor pressure.

Among the proceedings of ILIAS 6th Annual Meeting (Dresden, Germany, February 2009 [35]), there is a report of the second work group (WG2: Bank

of Isotopes, Sarazin [36]). In this report, the information is presented about a possible European facility for stable isotope enrichment, based on ICR-method: productivity up to 100 kg/year and device is flexible to produce most of the DBD isotopes. Documentation on the facility was prepared. Costs, time schedule and man power for a 3-4 years realization program was estimated.

The facility could be realized with a large current separator, ICR machine, and low current separator, Calutron. The project includes a chemical laboratory, a analytic laboratory of the atomic and isotopic composition of samples of matter (ICP, inductively coupled plasma discharge for atomic emission spectrometry, MS, mass spectrometers) and cryogenic support to ensure the installation by liquid nitrogen and liquid helium.

But no general agreements on this possible strategy was found in DBD community (large and expensive project).

Authors of the project are not presented in the publication. It is known that at the initial stage the developers of the project consulted with Louvet, former leader of experiments on ERIC installation [5].

2.3 Russia

In Russia, the technical project of the new ICR installation was ready in 2000 [25]. The main difference between the proposed construction and the design of PSP-installation (Fig. 4) was the fact that it was planned more than a high value of the magnetic field (4-Tesla). Authors of the project have conclusive experimental evidence that the isotopic selectivity of the cyclotron heating increases with increasing of magnetic field [6].

Preparation of the project was contributed by collaboration in the field of ICR isotope separation between IMPh (Institute of Molecular physics) of NRC "Kurchatov Institute" and Department of Enrichment Processes of Saclay Research Center (France). During the collaboration, taking place in the period of the

1990s, an opinion is occurred rather naturally about feasibility of further steps towards practical application of ICR method. The French side supported the Russian side in ISTC in creation of the project of prototype industrial installation. Researchers from Saclay became initially collaborators of the development. ISTC Project # 0830 [25] titled "Development of the project of the pilot installation MCIRI for separation of stable isotopes by method of ICR (ion cyclotron resonance) heating. Technical and economical substantiation of the project". It was carried out in 1999-2000 under leadership of Karchevsky (IMPh) and Keilin (ISSSPh-Institute of Superconductivity and Solid State Physics), NRC "KI". Technical and economical substantiation was made in the project institute VNIPIET in St. Petersburg. Enrichment of gadolinium and calcium by isotopes ¹⁵⁷Gd and ⁴⁸Ca had been considered for the purpose of mentioned above usage of enriched gadolinium as a burnable poison in the fuel of nuclear power reactors. Also another one considered task was mentioned above, where enriched calcium is required. Besides, calcium, enriched by ⁴⁸Ca isotope is required, in less amounts, for experiments at accelerators to obtain superheavy elements (Oganessian, Dmitriev [37]).

Calculated productivity and cost of the product indicate on expediency of MCIRI (method of cyclotron ion resonance of isotopes) creation. However, high cost of the project—more than 200 million rubles, is not allowed to realize it.

Therefore, in recent years (2009-2011), people carried out study of the sketch of ICR installation a smaller scale, that was able to separate only isotopes of elements of middle mass [38-40]. Basically, the project aims at the extraction of neutron-excess isotope 48 Ca, in order to ensure the experiments on obtaining of the superheavy elements [37]. If incorporated into the project installation options are realized, then it will be possible also to obtain isotopes 102 Pd, 150 Nd, separated from the neighboring isotopes in two mass ($\Delta M = 2$).

In the 2000s, experiments on Sirena installation

were continued, aimed at increase of extraction coefficient of the target isotope in the ICR method [41, 42]. Also they were continued theoretical researches in this direction [43].

Recall, that the extraction coefficient γ is the ratio of the quantity of a target isotope, settled at the collector of the product to the quantity of this isotope, transferred by plasma flow during the separation time.

Besides, participants of the ISTC #0830 project worked on the project improvements. These improvements are contained in two Russian patents. The author will present here only new variants of collector systems. Accroding to Karchevskii et al. [44], a collector system design is proposed with radial collector plates, intended for collection of a product at the end of a solenoid in a weak magnetic field region with divergent strength lines (Fig. 7).

Collector with radial plates is convenient because at the edge of the solenoid strength lines of the field lie in the plane of the plates. When placing of the collector system in diminished magnetic field region, it is possible to extend exposition duration and to increase separation coefficient [23, 24].

This patent [44] was one of the last works of Karchevskii, who led the work on the separation of isotopes in the plasma at the Institute of Molecular Physics. His life was tragically cut short in 2003 [45].

The principal difficulty in collector system design is cooling of thin screens. Judging by Fig. 6 [28], in experiments on Gd, Dy and Er isotope separation the screens did not cooled. Obviously, evaporation of matter from screens without cooling will affect on concentration of extracted isotope in the product. Only at separation of isotopes of some elements, having low vapor pressure at temperatures about 1,000 °C, for example, hafnium, screens without cooling can be used. The next problem: separate cooling of screens, product collectors and waste collector. Accroding to the patent of Gorshunov et al. [46], constructions with contact cooling of screens were proposed (through insulator with high thermal conductivity—for example SiC). A

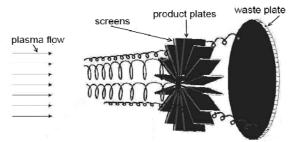


Fig. 7 Collector system for settling of heated ions in the magnetic field decreasing along the installation axis [44].

collector system in a form of pyramid, allow to cool the body frame only (Fig. 8). Step size L is selected approximately equal to the step of screw trajectory of heated ions in the given cross section of solenoid field, and height $d \approx H-h$.

In the recent years, a new approach in the field of plasma separation starts (Smirnov et al. [47, 48]), aimed at nuclear waste treatment. During nuclear reactor operation, products of nuclear fission (nuclear ashes), accumulating in nuclear fuel, impede chain reaction process. In the work of nuclear waste treatment, it is necessary, in particular, to separate a group of chemical elements, constituting nuclear ashes. It is proposed, to use ICR method for this purpose, transferring the matter into the plasma state. Heating of a group of ions, constituting nuclear ash, by monochromatic RF field, should be carried out in a non-uniform magnetic field. In this case, according to the ion cyclotron resonance condition, ions of different masses, will undergo cyclotron resonance in different cross sections of the heating zone. In NRC "KI", a plasma separator PS-1 was created. This installation must show on model mixtures a principal possibility of plasma technology usage in the process of nuclear waste treatment. In Fig. 9, a principal scheme is presented of "maximum" variant of the installation: toroidal separator [49] will not be used, if methods of settling, used at isotope separation will be effective. As a source of metallic plasma an ECR discharge in magnetic mirrors trap was applied.

Obstacle to the practical use of this scheme can be formation of multiply charged ions that can be originated either in transformation material into the

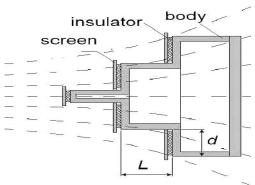


Fig. 8 Pyramidal collector system in non-uniform (flaring) magnetic field [46].

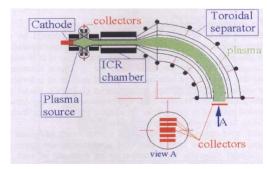


Fig. 9 Layout of plasma separator PS-1 [47].

plasma state or at a flight of plasma to the zone of separation [50].

3. Conclusions

Thus, people has not realized that there is no large-scale project to create an ICR-installation during the 2000s. The realization of such expensive projects (cost ~\$10 million) depends on the scale of the need for isotopes that can be obtained at the proposed facilities. Grossbeck et al. [28] suggest that in the future, when used for electricity production the reactors of IV Generation [51], the need for enriched dysprosium and erbium is such that would worth-while the creation of a large device of ICR. Recall, that originally planned to use the PSP in Oak Ridge for pre-enrichment of raw materials used in the calutrons [52].

More a simple problem—the extraction of isotopes 150 Nd ($\Delta M = 2$) and 48 Ca ($\Delta M \ge 4$) in order to observe neutrinoless DBD. Unknown fate of the project of European ICR facility [36] was after the completion of activities within ILIAS in 2009. Apparently, require the activity of the participants of DBD projects, such as

occurs in the project SuperNEMO [53]. But the activity of the participants of this project is not for the ICR method. In agreement with the Korea Atomic Energy Research Institute, there is the project extract isotope ⁴⁸Ca by laser AVLIS-method. People expected to make an order in Russia in the extraction of ¹⁵⁰Nd using AVLIS. The project of European ICR-facility is related to another research group of DBD (Cuore, Italy).

Continue theoretical research on ways to nuclear waste treatment by plasma ICR method (Timofeev [54]). The aim of this work—calculation of the number of multiply charged ions produced in the plasma moving from the source to the zone of selective heating of ions. This parameter is critical to the process of plasma treatment of spent nuclear fuel. The latter work allows to evaluate the possibilities of this method of treatment more optimistic than on the basis of Ref. [50]. This is important and to the process of isotope separation by ICR [9, 15, 55]. But it was discussed mainly about the loss of the isotopes: doubly charged ions are not heated at the cyclotron frequency of single charged and direct in the waste.

Do not discuss the influence of doubly charged ions on the selectivity of the cyclotron heating, although the frequency of collisions of singly charged ions with multiply $\sim Z^2$. At $\approx 15\%$ of multiply charged ions in the plasma (the part, which measurements showed by a probe mass-spectrometer in the gadolinium—argon plasma at the facility ERIC [15] and which is confirmed by evaluation [55]) will have a significant broadening of the ICR-lines. Perhaps the indisputable conclusion is given in Ref. [9]: at an electron temperature Te ~ 1 eV the proportion of doubly charged ions is small. If to bear in mind that for selective heating of ions of isotopes of rare earth elements, it is necessary that the ion temperature should be higher than 5 eV [29], then it is clear that the plasma with unusual parameters is required.

Is it possible to judge on the influence of multiply charged ions from the results of experiments? It is known that the ICR method successfully separated the isotopes of nickel [11], molybdenum, palladium [12]. These three elements of the second ionization potential is higher than that of gadolinium: 18.2, 15.7, 19.4 eV, respectively (U_2 (Gd) = 12.1 eV [56]). But there is another factor that facilitated the task in the case of these three elements: isotopes are separated from the next with the two masses (62 Ni, 100 Mo, 102 Pd; $\Delta M = 2$). The maximum separation factor, achieved in the ICR method, $\alpha \approx 130$ (isotopes of calcium [57], $U_2 = 11.9$ eV). But again, $\Delta M \ge 4$ (the concentration of the isotope 44 Ca $\sim 2\%$). So clearly, that on the basis of experiments the influence of multiply charged ions is not obvious. However, at the planning the separation of isotopes of gadolinium and other rare earth elements in the ICR device desirable to provide an allowance for the magnetic field. Consequently, out of the proposed 4-6 T, 6 T should be chosen.

The problems of multiply charged ions is absent at the separation of isotopes of alkali metals (Li, K and Rb), having a high potential for double ionization.

Universal ICR installation—an expensive device ("Expensive" [36]). The superconducting magnet, gyrotrons, the vacuum system—these are the main components of the high cost. The plan, for example, to replace the microwave discharge source by a discharge in crossed electric and magnetic fields (Volosov et al. [58]), giving gyrotrons up. There are other proposals to improve the ICR method, the subject of patent development. All of them are ultimately aimed at increasing the separation power of ICR systems and their implementation will accelerate the return on investment. Based on the above facts and estimates point out the options of a installation that can separate the isotopes of rare earth elements: $B \le 6 \text{ T}$, $Te \sim 1 \text{ eV}$, $Ti \ge 5 \text{ eV}$.

It should be considered whether to choose one—the only method for the separation of isotopes of the elements that have no chemicals with sufficient vapor pressure and the centrifugal method which does not apply. If you strive to meet present and future needs in the isotopes of these elements, it is advisable to have

the opportunity to apply an EM (electromagnetic), laser (AVLIS) or plasma (ICR) methods, depending on the specific task. Virtually, all of these isotopes used in practice, are obtained by EM-method. Its products are in demand now [59]. A combination of these methods, such as incorporated in the project [36]: ICR-EM may contribute to production increase.

Acknowledgments

This work was supported by Russian FBR grant (Project 11-02-12098-ofi-m).

References

- [1] J.M. Dawson, H.C. Kim, D. Arnush, B.D. Fried, R.W. Gould, L.O. Heflinger, et al., Isotope separation in plasmas by use of ion cyclotron resonance, Phys. Rev. Lett. 37 (1976) 1547-1550.
- [2] J.P.M. Schmitt, Dispersion and cyclotron damping of pure Bernstein waves, Phys. Rev. Lett. 31 (1973) 982-986.
- [3] G.A. Askarian, V.A. Namiot, A.A. Rukhadze, Change of plasma mass composition in magnetic trap by ICH, JTP Lett. 1 (1975) 356-360. (in Russian)
- [4] M. Mussetto, T.E. Romesser, D. Dixon, C. Strawitch, J. Tang, Isotope separation using selective ion cyclotron resonance heating in metallic plasmas, in: IEEE Int. Conf. on Plasma Science, San Diego, Calif., 1983, p.70.
- [5] P. Louvet, Review of isotopic plasma separation processes, in: Proc. of 2nd Workshop on Separation Phenomena in Liquids and Gases (SPLG), Versailles, France, 1989, Vol. 1, pp. 38-77.
- [6] A.I. Karchevskii, V.S. Laz'ko, Y.A. Muromkin, A.I. Mjachikov, V.G. Pashkovsky, A.L. Ustinov, et al., Investigation of lithium isotope separation in a plasma with isotopically selective ICR heating, Plasma Phys. Rep. 19 (1993) 214-222. (in Russian)
- [7] U.S. University of California (UCLA) Home Page, http://www.physics.ucla.edu/plasmalab/currActIndex.htm1.
- [8] Method and Apparatus for Isotope Separation, Applicant: TRW INC., US International Application published under the Patent Cooperation Treaty (PCT), July 19, 1984.
- [9] F.F. Chen, Double helix: The dawson separation process, in: T. Katsouleas (Ed.), From Fusion to Light Surfing: Lectures on Plasma Physics Honoring John M. Dawson, Addison-Wesley, New York, 1991, pp. 191-211.
- [10] T.E. Romessor, V. Vanek, J. Tang, D. Dixon, J. Bayless, A large area metallic ion plasma source, in: IEEE Int. Conf. on Plasma Science, San Diego, Calif., 1983, p.71.
- [11] Plasma Separation Process, Quarterly Technical report for

- Period 1/10/86 to 31/12/86, DOE/ET/33006-001, pp. 14-16.
- [12] N.R. Stevenson, T.S. Bigelow, F.J. Tarallo, Industrial scale production of stable isotopes employing the technique of plasma separation, Journal of Radioanalytical and Nuclear Chemistry 257 (1) (2003) 153-155.
- [13] P. Louvet, L. Metayer, The use of ion cyclotron resonance for separation of stable isotopes, in: Compte rendu des Journees sur les Isotopes Stables, Saclay, 1993 (Workshop on Stable Isotopes: Applications-Production), Le livre des communications, pp. 195-202 (Ed. par le CEA, 1994).
- [14] P. Louvet, A. Compant La Fontaine, B. Larousse, M. Patris, Heavy stable isotope separation by ion cyclotron resonance, in: Proc. of 4th Workshop on Separation Phenomena in Liquids and Gases (SPLG), Beijing, P.R. China, 1994, pp. 83-92.
- [15] A. Pailloux, A. Compant la Fontaine, P. Louvet, Separation of gadolinium isotopes by the ion cyclotron resonance process, in: 6th Workshop on Separation Phenomena in Liquids and Gases (SPLG), Nagoya, Japan, 1998, pp. 332-343.
- [16] A. Compant La Fontaine, P. Louvet, P. Le Gourrierec, A. Pailloux, Selective heating of high-pressure vapour elements by ion-cyclotron resonance, J. Phys. D: Appl. Phys. 31 (1998) 847-865.
- [17] A.I. Karchevsky, V.S. Lazko, Y.A. Muromkin, V.G. Pashkovsky, A.L. Ustinov, D.A. Dolgolenko, Enriched lithium collection from lithium plasma flow, in: Compte rendu des Journees sur les Isotopes Stables, Saclay, (Workshop on Stable Isotopes: Applications-Production), Le livre des communications, 1993, pp. 343-353.
- [18] D.A. Dolgolenko, A.I. Karchevsky, V.S. Laz'ko, Y.A. Muromkin, V.G. Pashkovsky, A.L. Ustinov, On some peculiarites of the ICR-method of isotope separation, in: Proc. of 4th Workshop on Separation Phenomena in Liquids and Gases (SPLG), Beijing, P.R. China, 1994, pp. 93-99.
- [19] D. Dolgolenko, A. Karchevsky, V. Laz'ko, Y. Muromkin, V. Pashkovsky, Measurements of the extraction coefficient at lithium isotope separation by ICR method, in: 6th Workshop on Separation Phenomena in Liquids and Gases (SPLG), Nagoya, Japan, 1998, pp. 344-350.
- [20] N.M. Gorshunov, D.A. Dolgolenko, G.E. Zotin, A.I. Karchevsky, V.S. Laz'ko, Separation of lithium isotopes in plasma by ion cyclotron resonance method, Prikladnaja Fizika (Applied Physics) N3 (2001) 34-40. (in Russian)
- [21] J.S. Carty, A. Wong, G. Rosenthal, Separation of rubidium isotopes by plasma method enrichment process, in: 4th Int. Conf. on Isotopes, Cape Town, South Africa, 2002, p. 36.
- [22] A.I. Karchevskii, Y.A. Muromkin, Isotope separation in plasma by selective ICRH, in: V.Y. Baranov (Ed.),

- Izotopy: Svoistva, Poluchenie, Primenenie (Isotopes: Properties, Production, Application), Fizmatlit, Moscow, 2005, Vol. 1, pp. 307-325. (in Russian)
- [23] D.A. Dolgolenko, A.I. Karchevsky, V.S. Laz'ko, Y.A. Muromkin, V.G. Pashkovsky, Study of Extraction of Enriched Lithium in a Weakened Magnetic Field, Reports of the 2nd All-Russian Scientific Conf. "Physical-Chemical Processes in Selection of Atoms and Molecules", TsNIIATOMINFORM, Moscow, 1997, pp. 94-97. (in Russian)
- [24] A.I. Karchevskii, E.P. Potanin, To the question about using a weakened magnetic field in order to increase the efficiency of ICR installation, Plasma Phys. Rep. 25 (1999) 273-276. (in Russian)
- [25] ISTC Project # 830 Summary Home Page, http://www.istc.ru/istc/db/projects.nsf/.
- [26] C. Haynam, B. Comaskey, J. Conway, J. Eggert, J. Glaser, E. Ng, et al., Status of Gadolinium Enrichment Technology at LLNL, The Society of PIE, 1993, pp. 24-36.
- [27] J.P.A. Renier, M.L. Grossbeck, Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors, Report on Phase 1 of NERI Project N 99-0074, U.S. Oak Ridge National Laboratory, 2001, p. 185.
- [28] M.L. Grossbeck, J.P.A. Renier, T. Bigelow, Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors, Final Report on NERI Project Number 99-0074, USA, Sept. 2003, p. 133 (Prepared by the University of Tennessee).
- [29] D.A. Dolgolenko, Y.A. Muromkin, Plasma isotope separation based on ion cyclotron resonance, Physics-Uspekhi 52 (4) (2009) 345-357.
- [30] Y.A. Muromkin, The status of the isotope separation by PSP, in: Proc. of 11th Workshop on Separation Phenomena in Liquids and Gases, Saint-Petersburg, Russia, June 13-18, 2010, pp. 104-113.
- [31] U.S. DOE Office of Science Web site, http://science.energy.gov/np/research/idpra/workshop-on-the-nations-needs-for-isotopes-present-and-future/.
- [32] U.S. DOE Office of Science, Report of A.Y Wong [Online], http://science.energy.gov/~/media/np/pdf/research/idpra/ Nonlinear%20Ion%20Dynamics%20LLC.pdf.
- [33] J.M. Dawson, Isotope separation by magnetic fields, US Patent, N4081677 (1978).
- [34] U.S. DOE Small Business Innovation Research Web site, http://www.sbir.gov/sbirsearch/detail/250566.
- [35] Agenda of ILIAS 6th Annual Meeting Web site, Dresden, Germany, 2009, http://ilias.in2p3.fr/ilias_site/meetings/agenda/ILIAS_6th Annual Meeting.htm.
- [36] E.U. Integrated Large Infrastructure for Astroparticle

- Science Web site, http://ilias.in2p3.fr/ilias_site/meetings/documents/ILIAS_6th Annual Meeting/170 209 Sarazin2.pdf.
- [37] Y.T. Oganessian, S.N. Dmitriev, Isotopes of transactinides and superheavy elements, in: V.Y. Baranov (Ed.), Izotopy: Svoistva, Poluchenie, Primenenie (Isotopes: Properties, Production, Application), Fizmatlit, Moscow, 2005, Vol. 2, pp. 45-62.
- [38] N.M. Gorshunov, V.S. Las'ko, Y.A. Muromkin, E.P. Potanin, A.L. Ustinov, Estimation of the parameters of the separative ICR installation for the extraction of neutron-surplus isotopes of calcium-48, Perspektivnie Materiali (Perspective Materials) 10 (2011) 72-79. (in Russian)
- [39] N.M. Gorshunov, D.A. Dolgolenko, Y.A. Muromkin, E.P. Potanin, A.L. Ustinov, ECR sources of calcium plasma, Instruments and Experimental Techniques 54 (1) (2011) 98-104.
- [40] O.P. Anashkin, A.S. Dinisilov, V.E. Keilin, I.A. Kovalev, A.V. Naumov, V.I. Sherbakov, The superconducting magnet for the installation on the isotope of calcium-48 to pick by ICR-method, Perspektivnie Materiali (Perspective Materials) 10 (2011) 116-121. (in Russian)
- [41] A.P. Babichev, N.M. Gorshunov, D.A. Dolgolenko, G.E. Zotin, A.I. Karchevsky, Research of neutral atoms flow in ICR installation for lithium isotope separation, in: Proc. of VII All-Russian (International) Scientific Conference "P-C P...", Zvenigorod, Moscow Region, TsNIIATOMINFORM, 2002, pp. 208-213. (in Russian)
- [42] V.G. Pashkovsky, Definition of flows of neutral atoms from different sources in the installation for lithium isotope separation, in: Proc. of VII All-Russian (International) Scientific Conference "P-C P..." Zvenigorod, Moscow Region, TsNIIATOMINFORM, 2002, pp. 214-219. (in Russian)
- [43] E.P. Potanin, Heating of gadolinium ions by ICR method, Tech. Phys. 51 (2006) 1586-1593. (in Russian)
- [44] A.I. Karchevskii, E.P. Potanin, Method for separating stable isotopes in plasma by ion-cyclotron resonance and device for its implementation, Russian Federation Patent, 2217223 (2003).
- [45] In Memory of Aleksandr Ivanovich Karchevskii, Plasma Physics Reports, 2004, Vol. 30, pp. 272-273.
- [46] N.M. Gorshunov, D.A. Dolgolenko, V.S. Laz'ko, Y.A. Muromkin, V.G. Pashkovsky, et al., Device for isotope separation in plasma by ion-cyclotron resonance (variants), Russian Federation Patent, 2278725 (2006).
- [47] V.P. Smirnov, V.A. Zhiltsov, A.I. Morozov, N.N. Semashko, A.A. Skovoroda, A.V. Timofeev, Plasma separation of used nuclear fuel and nuclear waste, in: Proc. of IX All-Russian (International) Scientific Conference, Zvenigorod, Moscow Region, TsNIIATOMINFORM,

- 2004, pp. 7-11. (in Russian)
- [48] V.A. Zhiltsov, V.M. Kulygin., N.N. Semashko, A.A. Skovoroda, V.P. Smirnov, et al., The possibility of application of plasma separation methods to work with nuclear materials, At. Energy 101(2006) 755-761. (in Russian)
- [49] A.V. Timofeev, Ion separation by a curved magnetic field in a multicomponent plasma, Plasma Phys. Rep. 26 (7) (2000) 626-627. (in Russian)
- [50] A.V. Timofeev, Plasma processing of spent nuclear fuel by two-frequency ion cyclotron resonance heating, Plasma Phys. Rep. 35 (11) (2009) 912-923. (in Russian)
- [51] A technology roadmap for generation IV nuclear energy systems, Dec. 2002, US, DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum [Online], http://www.ne.doe.gov/genIV/documents/gen_iv_roadma p.pdf.
- [52] J.G. Tracy, Isotope separation program—Present and future, Nucl. Instr. and Meth. in Phys. Res. A 282 (1989) 261-266.
- [53] F. Piquemal, A. Petit, Future needs in enriched isotopes for the SuperNEMO project, in: Proc. of 11th Workshop on Separation Phenomena in Liquids and Gases,

- Saint-Petersburg, Russia, June 13-18, 2010, pp. 65-70.
- [54] A.V. Timofeev, Flow of a plasma of multielectron elements along a magnetic field, Plasma Phys. Rep. 37 (11) (2011) 978-987. (in Russian)
- [55] D.A. Dolgolenko, A.I. Karchevsky, E.P. Potanin, Estimation of Concentration of the Single and Twice Charged Ions in Gadolinium Plasma Source, Reports of the V All-Russian (International) Scientific Conf. "P-C P...", TsNIIATOMINFORM, Moscow, 2000, pp. 130-133. (in Russian)
- [56] I.S. Grigor'ev, Y.A. Danilov, E.Z. Meilikhov, Tables of Physics Values, Hand-book, in: I.K. Kikoin (Ed.), Atomizdat, Moscow, 1976, pp. 419-420. (in Russian)
- [57] A.C. La Fontaine, C. Gil, P. Louvet, Separation des isotopes du calcium par resonance cyclotron ionique, C. R. Acad. Sci. Paris 308 (1989) 821-825.
- [58] V.I. Volosov, I.A. Kotel'nikov, I.N. Churkin, S.G. Kuz'min, A.G. Steshov, A.V. Timofeev, Device for isotope separation by the method of the ICR Heating, At. Energy 88 (2000) 385-393. (in Russian)
- [59] I.A. Kabanov, Production by the separation electromagnetic method of highly enriched stable isotopes, Perspektivnie Materiali (Perspective Materials), Special Issue (10) (2011) 86-92. (in Russian)