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ISOTOPE SEPARATION OF ^{22}Na AND ^{24}Na
WITH USING LIGHT INDUCED DRIFT EFFECT

1993

I. Introduction

After the Light Induced Drift (LID) was predicted theoretically [1] and confirmed experimentally for atomic and molecular gases [2, 3] many theoretical and experimental studies have been dedicated to the investigation of this effect (e.g. the references in [4]). Such interest in the LID effect is based on extended prospects of LID in scientific and technical applications. Isotope separation with using LID effect is one of them.

LID can occur when optically absorbing atoms immersed in a buffer gas are velocity-selective excited. Assume the exciting laser radiation is slightly detuned from the transition frequency of the absorbing atoms and only the atoms with a Doppler shift compensating the detuning interact with laser radiation. As a result the excited atoms move in one direction and the non-excited atoms move predominantly in the opposite direction. When the flux movements take place in a buffer gas the fluxes feel the collision resistance. Since gas kinetic cross sections for ground-state and excited atoms with buffer gas particles are different, the collision resistance forces for the excited and non-excited atoms fluxes also differ. Consequently, the whole absorber gas is driven by the buffer gas, providing the drift of the absorber atoms.

The optical resonance wavelengths for absorbers containing different isotopes are usually slightly shifted relative to each other. It enables to tune the laser radiation wavelength in such a way that the LID fluxes of absorbers with different isotopes have different (even opposite) velocities. This effect can produce a separation of the absorbers and, consequently, their isotopes. The isotope separation with using LID called shortly LIDIS (Light Induced

Drift Isotope Separation) is discussed in [5]. It has been investigated experimentally for some pairs of natural isotopes: $^{85,87}\text{Rb}$ with LID of atoms [6] and $^{12,13}\text{C}$, $^{14,15}\text{N}$, $^{79,81}\text{Br}$, $^{16,18}\text{O}$ and $^{32,34}\text{S}$ with LID of molecules [7].

LIDIS is a suitable method for radioactive isotope and isomer separation. The first experimental LIDIS investigation for radioactive isotopes was performed with ^{22}Na and ^{24}Na [5]. There was large discrepancy between the theoretical enrichment ($E > 100$) of the using LIDIS separator and the experimental results ($E = 0.11 \pm 0.04$, 0.19 ± 0.027). The present paper reports the new results of our investigations.

In Sec. II we give the discussion of the LIDIS separation with influence of molecular form. In Sec. III we describe the separation of the radioactive isotopes $^{22,24}\text{Na}$ on the LIDIS separator with improved buffer gas purification and discuss the experimental results.

II. Theoretical approach

We confine discussion to the LIDIS separator used in our experimental investigations. It is illustrated schematically in Fig. 1. The main part of the separator is a separation pipe. The sample of a mixture of two separated isotopes a and b is located in the separative pipe center. Atoms of the isotopes are released by thermal evaporation. The laser beam pass through the separative pipe. The laser wavelength is tuned in such a way that drift velocities of separated isotopes have the same value but opposing direction ($U_a = -U_b$). For isotope collection there are collectors in the separative pipe.

In the general case we must take into account the motions of the separating isotopes in the different physical and chemical states (atoms, ions, molecules) in the buffer gas, the effects of the surface adsorption and diffusion

both on the separative pipe so on the collector, the diffusion in the separative pipe material and adsorbing capability of the collector. All the processes can be described by three-dimension diffusion equation system.

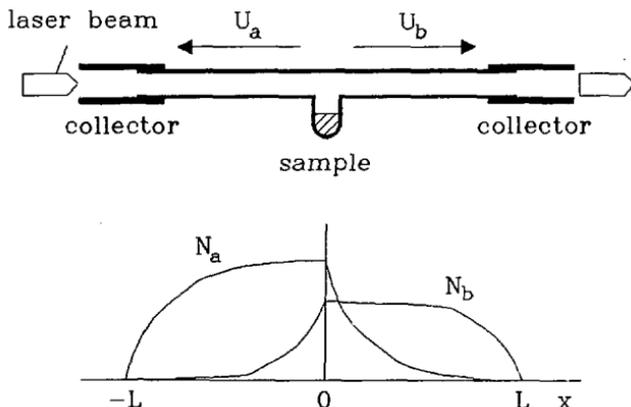


Fig. 1. Scheme of the LIDIS separation of isotopes A and B. The density of the separated isotope atoms $N_a(x)$ and $N_b(x)$ in separation pipe when $|U_a|L/D_a = |U_b|L/D_b = 5$.

When the condition $\phi \ll L$ (where ϕ is the inner diameter of the separating pipe, L is the distance between the sample and the collector) is fulfilled the LIDIS separator can be described correctly in one dimension approximation.

$$\partial N_i^k / \partial t = Q_i^k + \partial^2 D_i^k N_i^k / \partial x^2 - \partial U_i^k N_i^k / \partial x - \gamma_i^k N_i^k + \sum_l \beta_i^{kl} N_l^l, \quad (1)$$

$$\gamma_i^k = 1/\tau_i^k,$$

where the indexes of i determine the isotope, k and l determine the physical or chemical states (in the buffer gas, on the surface, in the solid material of the separative pipe and collectors) of the given isotope, Q_i^k is a source of the

i isotope in the k state, D is the diffusion coefficient, N_i is the given isotope density, U_i^k is LID velocity ($U_i^k \neq 0$ only for free neutrals atoms), τ_i^k is the lifetime of the given isotope in the k state, β_i^{kl} is the transition probability of the i isotope from the l state to the k state.

Suppose the N_i^k distribution in the separative pipe is stationary, the drift velocities and the diffusion coefficients are constant along the separation pipe. Suppose that atomization of the sample goes with a certain efficiency (only a part of the isotopes is in the state of the free atoms). and there is no transitions between the physical or chemical states $\gamma_i^k = 0$ $\beta_i^k = 0$. In such case the equation system (1) can be simplified to

$$D_i^k \partial^2 N_i^k / \partial x^2 + U_i^k \partial N_i^k / \partial x = 0, \quad (2)$$

Suppose $x = 0$ is the separation pipe center, the boundary conditions are $N_i^k(0) = N_{0i}^k$, $N_i^k(L) = 0$, the diffusion coefficients have an equal value for both isotopes ($D_a^k = D_b^k = D^k$). Then the $N_i^{at}(x)$ distribution of free atoms of separated isotopes in the separation pipe is given by

$$N_i^{at+}(x) = N_{0i}^{at} \left\{ 1 - \exp \frac{-U_i(L - |x|)}{D^{at}} \right\}, \quad (3)$$

$$N_i^{at-}(x) = N_{0i}^{at} \exp(-U_i |x| / D^{at}), \quad (4)$$

where $N_i^{at+}(x)$ and $N_i^{at-}(x)$ are the atom density increased and decreased by LID respectively. The examples of $N_i^{at}(x)$ distribution of atoms are given in Fig. 1.

The j_i^{at+} and j_i^{at-} fluxes of the separated isotopes on the collectors increased and decreased by LID, respectively, are given by

$$j_i^{at+} = U_i N_{0i}^{at}, \quad (5)$$

$$j_i^{at-} = U_i N_{0i}^{at} \exp(-U_i L / D^{at}). \quad (6)$$

Such fluxes give the following "theoretical" separation factor α_i^{th} on the collectors

$$\alpha_i^{th} = \exp(U_i L / D^{at}). \quad (7)$$

Let us substitute real experimental value for sodium atoms and argon systems in [11]. If $|U_i| = 100$ cm/s, $L = 3$ cm and $D^{at} = 20$ cm²/s then $\alpha_a^{th} \approx 3 \cdot 10^6$. An example of the dependence of the α_a separation factor on the separation pipe length L is given in Fig. 2. Unfortunately the enrichment can be spoiled by the isotopes which are in the separating pipe in a molecular state. The "real" separation factor α_a^r on the collector to which the LID of a isotope atoms are directed is defined by

$$\alpha_a^r = \frac{\int (j_a^{at+} + j_a^{mol}) dt}{\int (j_b^{at-} + j_b^{mol}) dt} \cdot (N_{0a}^{at} / N_{0b}^{at})^{-1}, \quad (8)$$

where the j_a^{at+} , j_a^{mol} , j_b^{at-} , j_b^{mol} are the atom and molecule fluxes of the a and b isotopes on the collector. The "real" separation factor α_b^r is defined analogously as α_a^r in (8). If $j_i^{at} / j_i^{mol} = \text{constant}$ and $j_i^{at-} \ll j_i^{mol}$ then (8) can be simplified to

$$\alpha_a^r = \frac{j_a^{at+} + j_a^{mol}}{j_b^{mol}} \cdot (N_{0a}^{at} / N_{0b}^{at})^{-1}. \quad (9)$$

If $j_a^{mol} / N_{0a}^{at} = j_b^{mol} / N_{0b}^{at}$, then (9) can be simplified to

$$\alpha_a^r = j_a^{at+} / j_a^{mol} + 1 = 2j_a^{at} / j_a^{mol} + 1 = 2\vartheta + 1, \quad (10)$$

where j_a^{at} and j_a^{mol} are the fluxes of the atoms and molecules

of the separating isotope element from the sample, ϑ is atomization factor of separating isotopes fluxes from the sample ($\vartheta = j_{\text{at}}^{\text{at}}/j_{\text{mol}}^{\text{mol}}$). The examples of the dependencies of the α_a^r separation factor on the separation pipe length L and on ϑ are given in Fig. 2.

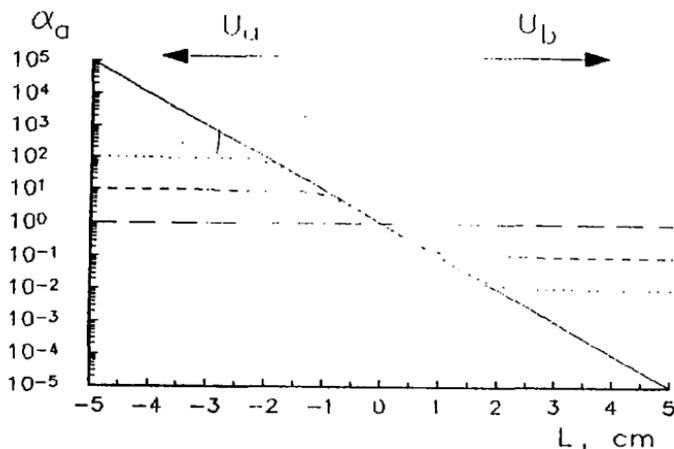


Fig. 2. Dependencies of the α_a separation factor on the length L of the separation pipe. The solid line represents theoretical separation factor α_a^{th} when $U/D = 2.3 \text{ cm}^{-1}$. The dots, short dash and long dash lines represent real separation factors α_a^r when the relations ϑ of atoms and molecules of the separated isotopes in the separation pipe center are 50, 9, 0 respectively.

III. Experiment and results

The ^{22}Na and ^{24}Na isotopes are very suitable for LIDIS experimental investigations for the following reasons. Firstly, the LID of stable sodium atoms is well investigated [6], the optimum experimental conditions and the corresponding LID velocity are known. Secondly, ^{22}Na and ^{24}Na are suitable isotopes for the LIDIS investigations. Their isotope shifts for the D_1 line are -756.9 and 706.0 MHz, respectively [8]. Laser radiation wavelength tuning at the center of the ^{23}Na line produces the LID of ^{22}Na and ^{24}Na atoms in the opposite directions [5]. Both LID velocities of the ^{22}Na and ^{24}Na atoms are near the maximum velocity that can be reached at the optimum laser wavelength tuning [9, 10].

A scheme of the experimental set-up is given in Fig. 3.

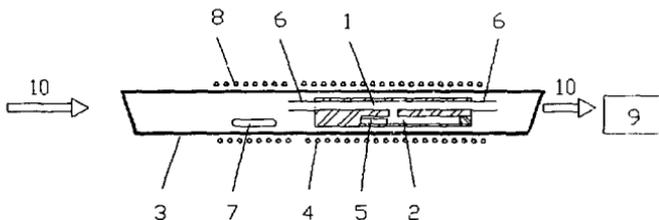


Fig. 3. Scheme of the LIDIS separator. The figure in the picture represents: 1, separative pipe; 2, quartz jacket; 3, pocket; 4, aluminum sample; 5, quartz collectors; 6, metallic potassium; 7, 8, heaters; 9, power meter; 10, laser beam.

A stainless separation pipe (1) with length of 2 cm and with inner diameter of 4 mm is fixed in a quartz jacket (2). The center of the separation pipe is connected with a pocket (3). The pocket contains 10 mg aluminum (4) with about 10^{12} atoms of ^{22}Na ($T_{1/2} = 2.62$ y) and about 10^9 atoms of ^{24}Na

($T_{1/2} = 14.96$ h). The isotopes had been produced by the aluminum spallation in a 660 MeV proton beam. At the ends of the separation pipe (1) there are two quartz collectors (5) which bind sodium atoms at high temperatures. An ampoule (6) containing metallic potassium is put at the collector nearer the laser. The separation pipe (1) and the ampoule with potassium are heated by separate heaters (7), (8).

The quartz jacket (2) and the separation pipe (1) were baked for 2 - 4 h at 850 K in vacuo $10^{-3} - 10^{-4}$ torr. Contemporarily the ampoule with potassium was heated up and a potassium mirror appeared on the quartz jacket (2). Then the quartz jacket (2) has been filled by 10 torr of a pure krypton (99.999 %). The beam (10) of a cw ring dye laser (Spectra Physics 380 D) with power of ≈ 40 mW passed through the separation pipe (1). The laser radiation wavelength was tuned to the D_2 line center of the ^{23}Na atomic spectrum ($\lambda = 588.99$ nm). The temperature of the potassium heater (8) was held at the level when in the heating region a yellow cloud was shining. It was caused by admixture of sodium atoms fluoresced in the laser beam but did not attenuate one. That guaranteed a high density of potassium atoms needed for effective buffer gas purification from admixture which could bind sodium atoms (O_2 , H_2O , and so on).

The separation pipe (1) was heated to above the aluminum melting point (933 K). At this temperature the time of the sodium atom adsorption on the stainless surface is $10^{-4} - 10^{-5}$ s and sodium in the separating pipe is moving [11]. Sodium atoms evaporate from the liquid aluminum and diffuse into the separating pipe. LID swept the ^{22}Na and ^{24}Na atoms in the separation pipe to the left and right respectively (see Fig. 1.) and produced $^{22}\text{Na}/^{24}\text{Na}$ enrichment on the left/right collectors. A power meter (9) measured the power of the laser beam transmission through the separating pipe (1). The density of the sodium atoms in the separation pipe was maintained in an optical thin range by a temperature control. The power of the laser beam passing through the separation pipe was kept above 20 mW. We assume that the laser beam was attenuated predominantly by the ^{23}Na

atoms which were present in a larger amount than the $^{22,24}\text{Na}$ atoms in the aluminum sample 4. The separation was performed for 1 h. Then the heater (7) was switched off and quartz jacket with separation pipe was cooled. Then the ^{22}Na and ^{24}Na activities (γ - line energy: 1.275 MeV for ^{22}Na and 1.369 MeV for ^{24}Na) on the collectors 5 were measured by a germanium detector with multichannel analyzer.

On the collectors there was collected up to 50 % of the sample activity.

Let us compare the theoretical and real characteristics of the separator at the conditions of this experiment.

The separation factor α_{22}^c of ^{22}Na can be expressed as

$$\alpha_{22}^c = \frac{A_{22}^c A_{24}^s}{A_{24}^c A_{22}^s} \quad (11)$$

where A_i^s and A_i^c are indicated isotopes activities in the sample and on a collector respectively at a certain time. In our experiment the ^{22}Na separation factors α_{22}^c were 17.85 ± 1.37 and 0.22 ± 0.012 for left and right collectors, respectively (Fig. 4). If all sodium was in atomic form, and $|U_{22}| = |U_{24}|$, then according to (7) $UL/D^{at} = 2.88$. In this experiment $L = 2$ cm the laser beam intensity $\approx 150 + 250$ mW/cm² the buffer gas pressure ≈ 10 torr and the dependence of the LID velocity on the temperature, the experimental value $U/L = 1.44$ cm⁻¹ is in agreement with analogous one ($U/D \approx 1.4$) deduced from experimental results in [12].

There was a hypothesis that sodium can diffuse from one collector to another between the separative pipe and quartz jacket. In consequences of this process the separation factor on the collectors is limited. For the effect elimination the following experiment was realized.

The sample was located directly into the stainless separation tube with 24 cm length and 3 mm inner diameter. The separation pipe ends were out of the heater (6) and

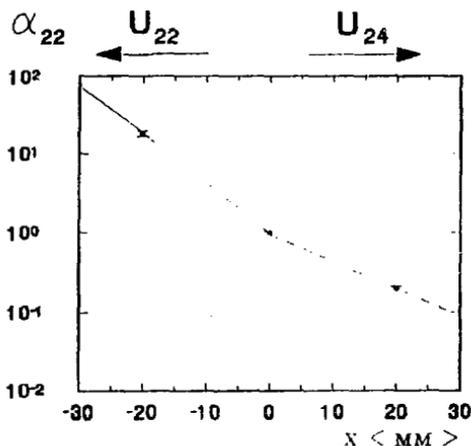


Fig. 4. The dependence of the experimental separation factor α_{22}^r of ^{22}Na on the separation pipe length. The solid and dashed lines represent the theoretical separation factors in the left and right parts of the separation pipe when $U_{22} = U_{24}$ and $U_{22}/D = 1.44$ and 0.75 cm^{-1} respectively.

their temperature was under 800 K. At that temperature the adsorption time of sodium atoms on a stainless surface is long and stainless can serve as a collector. The pressure of the buffer gas (pure krypton) was ≈ 10 torr. The power of the laser radiation passing through the separation pipe did not decrease under 15 mW during the separation process. Those experimental conditions correspond with theoretical separation factor $\approx 8 \text{ cm}^{-1}$. The separation experiment was performed for 0.5 h by the analogous way as in the first case. After that the separation pipe was extracted from the quartz jacket and cut into sections and the ^{22}Na and ^{24}Na activities in the sections were measured. Almost 50% of the original activity was in the sections which were 5 - 7 cm

from the source. There was no peak of 1.369 MeV (^{24}Na) on the γ -spectrum background of the left collector (section of 5 - 7 cm from the source). The about 45 % of the original activity remained in the source. The separation factors on the collectors were: $\alpha_{24} = 25.3 \pm 1.9$ and $\alpha_{22} \approx 20$ with reliability 96% (see Fig. 5.). The separation factors on the other separation pipe sections were lower than 5.

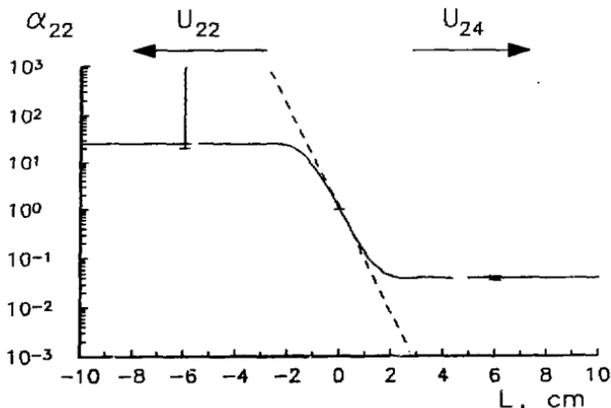


Fig. 5. The dependence of the experimental separation factor α_{22}^r of ^{22}Na the separation pipe length. The dash and solid lines represent theoretical and supposing real separation factors in the separation pipe under the given experimental conditions.

IV. Conclusions

The buffer gas purification and constructive modification allowed to improve the basic characteristics of the sodium LIDIS separator: separation factor and effectivity reached values of 25 and 50 %, respectively.

The most perspective methods in which the sodium isotope enrichment can be increased are: 1) A buffer gas flow through the separating pipe as it is described in [5]. 2) A separating pipe heating above the decomposition temperature of molecules with sodium atoms. 3) A molecule decomposition by an electron bombardment in the separating pipe. The last two ways are very perspective because they can give a possibility to use compounds as a sample of isotopes for separation and to separate isotopes of little volatile elements.

The separation efficiency can be increased from 50 % by extending of the separation time.

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Разделение изотопов ^{22}Na и ^{24}Na

с использованием эффекта светоиндуцированного дрейфа

В работе рассмотрен ЛИДИС (Light Induced Drift Isotope Separation) сепаратор без газового потока. Показано, что степень атомизации разделяемых изотопов ограничивает реальный коэффициент разделения. Улучшенная очистка буферного газа позволила увеличить экспериментальный фактор разделения изотопов ^{22}Na и ^{24}Na до 25. Новая экспериментальная установка позволила увеличить эффективность разделения до 50%.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна, 1993

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Isotope Separation of ^{22}Na and ^{24}Na
with Using Light Induced Drift Effect

The LIDIS (Light Induced Drift Isotope Separation) separator without a gas flow is discussed. It is shown, that atomization degree of the separated isotopes limited real separation coefficient. The better buffer gas purification allowed to increase the experimental separation factor of ^{22}Na and ^{24}Na isotopes up to 25. The new experimental set up allow to increase the separation efficiency up to 50%.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

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