Calcium isotope enrichment by means of multi-channel counter-current electrophoresis for the study of particle and nuclear physics

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We have developed a new method for enrichment of large amounts of calcium isotopes for the future study of \textsuperscript{48}Ca double beta decay. The method is called multi-channel counter-current electrophoresis (MCCCE). We present the concept of MCCCE, in which the power density in the migration path is the key to efficient enrichment of a large amount of materials. In MCCCE, ions migrate in multi-channels on a boron nitride (BN) plate, allowing a substantial increase in the power density to be achieved. We made a tiny prototype instrument and obtained an enrichment factor of 3 for the ratio of the abundance of \textsuperscript{48}Ca to \textsuperscript{43}Ca over that of natural abundance. This corresponds to an enrichment factor of 6 for \textsuperscript{48}Ca to \textsuperscript{40}Ca. This remarkably large enrichment factor demonstrates that MCCCE is a realistic and promising method for the enrichment of large amounts of ions. It can be applied to many other elements and compounds.

Subject Index D50, C44

1. Introduction

1.1. Enrichment in particle and nuclear physics

In particle and nuclear physics experiments, specific isotopes are sometimes vital. They are used as beams or targets in accelerator experiments. For instance, \textsuperscript{48}Ca is intensively used for studies of unstable nuclei and superheavy elements. For these studies, certain amounts of grams of isotopes with high enrichment are needed. Recently, further large amounts of enriched isotopes were required for the study of, for instance, double beta decay. Approximately 10 kg of highly enriched \textsuperscript{76}Ge was used by the Heidelberg–Moscow and IGEX experiments, and now the GERDA and Majorana experiments are using larger amounts. Recently, \sim 100 kg or more of \textsuperscript{136}Xe has been intensively used by the EXO and KamLAND experiments. The recent progress of the research can be found elsewhere [1]. Currently, the most cost-effective method of enrichment is centrifugation. Since this is applicable to gas, only elements that have gaseous compounds are enriched. Experiments with large amounts of enriched isotopes are being carried out only for such elements.
1.2. $^{48}\text{Ca}$

Among many isotopes, $^{48}\text{Ca}$ is very special. It is 20% heavier than $^{40}\text{Ca}$, which is the most abundant stable calcium isotope. Such a highly neutron-efficient isotope is used for many applications. However, its natural abundance is only 0.187%. $^{48}\text{Ca}$ is enriched by a mass separation method since calcium has no gaseous compound. The method could give enrichment higher than 90%. However, its cost is quite high, and thus one can only afford a small amount of, for instance, a gram or less.

$^{48}\text{Ca}$ is also quite attractive for the study of double beta decay, since it has the highest $Q$-value among all double beta decay nuclei. For the design of next-generation double beta decay experiments, development of a method to enrich $^{48}\text{Ca}$ at a kilogram-to-ton amount with reasonable enrichment is highly desirable. However, no cost-effective methods are available so far. Currently, CANDLES (T. Kishimoto et al., manuscript in preparation, and Refs. [2–4]) is an almost unique experiment, studying $^{48}\text{Ca}$ double beta decay with an appreciable amount of $^{48}\text{Ca}$. The good optical properties of the calcium fluoride (CaF$_2$) crystals used for the experiment allows the construction of a big detector. Thus, a substantial amount of $^{48}\text{Ca}$ can be prepared even for calcium of natural abundance. However, enrichment of $^{48}\text{Ca}$ is vital for the future study. The CANDLES Collaboration is developing cost-effective enrichment methods, for instance, the crown ether resin method (S. Umehara et al., manuscript in preparation). This method can process large amounts at low cost, in principle, although further improvement is still needed for practical operation.

Once enriched $^{48}\text{Ca}$ is available at relatively low cost, study of $^{48}\text{Ca}$ double beta decay could become a most attractive experiment. There have been many attempts to enrich $^{48}\text{Ca}$. However, no method has been successful for the cost-effective enrichment of large amounts of $^{48}\text{Ca}$.

2. MCCCE

2.1. CE

We developed multi-channel counter-current electrophoresis (MCCCE) for the effective enrichment of large amounts of materials. Electrophoresis has been used to separate isotopes. It employs the velocity difference of ion species moving under an electric field in water or liquid. It is well established and its principle is simple. In the following, we briefly describe some properties of the electrophoresis relevant to the present argument, since we do not intend to review a vast amount of works.

Usually, the velocity difference between isotopes is small and a long migration length is necessary to give an appreciable separation in distance for effective enrichment. Counter-current electrophoresis (CCE) was developed to realize an effectively long migration length for a given length of an instrument [5]. The enrichment demonstrated, however, was not enough for the realistic enrichment of large amounts of materials.

Capillary electrophoresis (CE) is widely used [6]. It employs a thin ($\sim 0.1$ mm$\phi$ or thinner) tube with a length of typically $\sim 1$ m. Migration under a high electric field ($\geq 100$ V/cm) gives clear separation within a short time (hundreds of seconds). It is used not only for the separation of elements but also much larger molecules like proteins. Although clear separation (enrichment) is obtained, CE is suitable only for the separation of tiny amounts of ions due to the small radius of the tube.

2.2. CCE with large diameter

Enrichment of a large amount of $^{48}\text{Ca}$ was attempted by CCE using a column of 2 cm diameter, which is much larger than that of CE [7]. About 22% increase in the enrichment factor (the ratio of
the abundances of \(^{48}\text{Ca}\) to \(^{40}\text{Ca}\) was achieved by migration of 23.6 m in 896 hours. Since the diameter is 2 orders of magnitude larger than that of CE, the cross-sectional area was increased by 4 orders of magnitude. However, the applied low electric field (\(\sim 1\ \text{V/cm}\)) resulted in a long migration time.

CE gives good separation in a short time, although it can process only tiny amounts. On the other hand, CCE could handle a large amount, but the separation was not so good, even after a long migration time. In the following, we would like to suggest that MCCCE can solve the shortcomings of both methods and make the enrichment of large amounts of materials possible.

2.3. Figure of merit of electrophoresis

Let us discuss the issues for efficient enrichment of ion species by electrophoresis. Under a certain electric field \(E\), electrically charged ions migrate a certain distance \(\ell\) within time \(t\), given as

\[
\ell = \mu E t,
\]

where \(\mu\) is electric mobility, which depends on the ion species. Here we take \(^{40}\text{Ca}\) and \(^{48}\text{Ca}\) as examples. By using the mobility difference between \(^{40}\text{Ca}\) and \(^{48}\text{Ca}\) as \(\Delta \mu = \mu(^{40}\text{Ca}) - \mu(^{48}\text{Ca})\), the difference in migration distance \(\Delta \ell\) appears as

\[
\Delta \ell = \Delta \mu E t \geq \sigma,
\]

where \(\sigma\) is the width of broadening of the migration distance. The enrichment becomes effective when \(\Delta \ell\) is larger than \(\sigma\). Thermal diffusion of atoms (\(\sigma_{\text{th}}\)) inherently exists for the broadening. The broadening due to thermal diffusion is given by a Gaussian distribution whose \(\sigma_{\text{th}} = \sqrt{2Dt}\), where \(D\) is the diffusion constant. For an ideal instrument where only thermal diffusion contributes, Eq. (2) gives

\[
t \geq \frac{2D}{(\Delta \mu E)^2}.
\]

In reality, the broadening (\(\sigma\)) includes not only \(\sigma_{\text{th}}\) but also other sources represented by \(\sigma_{o}\) due to, for instance, inhomogeneities of flow, current density, electric field, and so on. We can assume that the relation \(\sigma^2 = \sigma_{o}^2 + \sigma_{\text{th}}^2\) holds and \(\sigma_{o} \propto \sqrt{t}\) since it is also due to random motion. Then Eq. (3) holds by using effective \(D\) larger than that of the diffusion constant. The power density \(p\) is given by

\[
p = iE = \rho \mu E^2,
\]

where electric current density \(i = \rho \mu E\) and \(\rho\) is the ion concentration in solution. Thus, the time needed to achieve a certain separation given by Eq. (3) is inversely proportional to \(p\) or \(E^2\).

A higher electric field or power density gives a shorter time to achieve certain enrichment. A high power density can be tolerated in a very thin migration path like CE, though it can process only small amounts of ions. The question here is how we can increase the power density without sacrificing the cross-sectional area for the migration.

2.4. Cooling

The power density is limited by temperature, which is given by the balance of heating and cooling. The power generated in the channel escapes from its surface as heat flux. The heat flux density \(J\) is given by

\[
J = -\lambda \ \text{grad} \ T,
\]

where \(\lambda\) is thermal conductivity and \(T\) is temperature.
Here we consider two concentric cylinders, uniform in the $z$ direction, where the inner (outer) circle has the radius $r_i$ ($r_o$) and the thermal conductivities of their materials are $\lambda_i$ ($\lambda_o$) to estimate the temperature rise. The inner circle is for the migration path and the outer circle is for the insulator.

Joule heat is uniformly generated inside $r_i$ with a power density of $p$. The temperature at radius $r_o$, represented by $T(r_o)$, is kept constant by a cooling system. By applying Eq. (4), we obtain the following well known equation for the temperature rise $\Delta T = T(0) - T(r_o)$ as

$$\Delta T = \frac{pr_i^2}{4\lambda_i} \left( 1 + \frac{\lambda_i}{\lambda_o} \ln \left( \frac{r_o}{r_i} \right) \right).$$

(5)

The temperature at the center of the path $T(r_o) + \Delta T$ must be below 100 degrees.

The thermal conductivity of water $\lambda_i \sim 0.6$, which is the same for any instrument that uses aqueous solutions. Thus, Eq. (5) combined with Eq. (3) tells us that $pr^2$ or $r^2/t$ is almost constant for a given temperature rise. An increase in the cross-sectional area reduces the power density or prolongs the processing time. Roughly speaking, the amount of ions processable in a certain time is almost independent of radius for a single migration path, and the tolerated total power is almost the same.

2.5. **MCCCE**

MCCCE was invented to satisfy both large cross-sectional area and high electric field. It is realized by making many migration paths in an insulating material with large thermal conductivity. The migration paths have to be made in insulating materials to apply the electric field. Electrons carry electric current and heat in electrical conductors. Therefore, an insulator usually has low thermal conductivity. However, materials like BN (boron nitride), AlN (aluminum nitride), diamond, and so on do not follow this rule. They are insulators although they have large thermal conductivities because lattice vibration carries heat. The thermal conductivity of water and typical insulating materials used for a tube is around 0.6. On the other hand, the thermal conductivity of BN ($\lambda_{BN}$) could be 2000 at maximum and a BN plate with $\lambda_{BN} = 50$–100 is easily available. The use of such high-$\lambda$ material makes many migration paths possible. The size of the migration path is 0.8 mm $\phi$, as shown in the following: it is a channel rather than a capillary. We thus call the system multi-channel counter-current electrophoresis, abbreviated to MCCCE.

3. **Experiment**

3.1. **Instrument**

Figure 1 schematically shows the instrument that we used for the present experiment. It is made of acrylic resin and has an outer size of 8 cm in diameter and 13 cm in height. The size of the migration column is 4 cm in diameter and 9 cm in height. The essential part of the instrument is a migration path made in a boron nitride (BN) plate with dimensions of 80 mm $\phi$, 10 mm thick. The migration path consists of 69 channels (holes) of 0.8 mm $\phi$ every 4 mm spacing in a 4 cm diameter region. The thermal conductivity of BN ($\lambda_{BN}$) is 63, which is 2 orders of magnitude higher than that of water and the polyethylene or glass usually used for a tube. A BN plate with much higher $\lambda_{BN}$ is available, but this value is good enough to demonstrate that MCCCE works. We have an additional 5 mm thick BN plate, which has holes of 2 mm $\phi$ every 4 mm below the 10 mm BN plate. This is to reduce turbulence in the aqueous flow and for further removal of heat.
3.2. Temperature rise in BN

The dimensions of the channels in the BN plate determine the temperature rise for a given power density. The temperature rise in the 0.8 mmΦ channel \( T_{\text{ch}} \) is obtained by Eq. (5) without the second term. In order to give the temperature rise in the BN plate \( T_{\text{BN}} \), \( p \) is replaced by an average power density \( p_{\text{BN}} = p \frac{\pi 0.4^2}{4} = 0.031 p \), which includes the ratio of the area of a channel to the corresponding BN area. Other parameters are \( r_i = 2 \text{ cm}, r_o = 4 \text{ cm}, \lambda_i = 0.6 \), and \( \lambda_o = 63 \). These values give

\[
\Delta T_{\text{ch}} / \Delta T_{\text{BN}} \sim 1.3.
\]

This value tells us that the temperature rises in each channel and the BN plate are similar. A smaller value gives a larger power density for a given total temperature rise. However, a value close to unity is good enough, since room for further improvement is twice. On the other hand, we are discussing the order-of-magnitude improvement by MCCCE. Here, we consider heat leak only in the radial direction of the BN plate. In reality, the top and bottom surfaces contribute. Thus, this ratio is a guideline for operation of the instrument. We can provide a similar electric field to that in CE to the channel (migration path).

3.3. Operation

We are interested in operation that continuously produces a \(^{48}\text{Ca}\)-enriched solution with a fixed enrichment, since our goal is a realistic enrichment of a large amount of \(^{48}\text{Ca}\). CaCl\(_2\) solution is...
injected from inlet A, flows upwards, and is extracted from outlet B. The electric field forces the Ca ions to move downward. The migration speed of $^{40}$Ca due to the electric field is a little faster than that of $^{48}$Ca. When the parameters are optimized, $^{48}$Ca ions move upward faster than $^{40}$Ca ions. As a result, one expects a $^{48}$Ca-enriched solution from outlet B. At the beginning of operation, the calcium concentration of the extracted solution is very dilute and its enrichment of $^{48}$Ca might be higher (for the first few minutes), but our instrument is not optimized to measure such time dependence.

4. Results

4.1. Experimental results

In the experiment, we fixed the counter-current speed of the aqueous solution and changed the voltage to determine the migration speed. The speed of counter-current flow was set at 0.53–0.70 mm/s, which corresponds to an electric field of 90–120 V/cm, since the mobility of calcium ions is known to be 0.59 [mm/s]/[100 V/cm]. The CaCl$_2$ solution was 0.01 mol/l.

Table 1 shows $A_{43}/A_{48}$, which is the ratio of the abundance of $^{48}$Ca ($A_{48}$) to that of $^{43}$Ca ($A_{43}$). The abundances were measured by the inductively coupled plasma (ICP) mass spectrometer in the RI Center of Osaka University. The ICP mass spectrometer has difficulty in measuring $^{40}$Ca, since the argon gas used for the ion source obscures the signals of $^{40}$Ca. In contrast, the measurement of $^{43}$Ca is reliable. Thus, we measured $A_{43}/A_{48}$ and derived the enrichment factor $\alpha(48/43)$, which is the ratio of $A_{48}/A_{43}$ for a certain applied voltage and that for natural abundance. Figure 2 and Table 1 show $\alpha(48/43)$ and $\alpha_{est}(48/40)$, which are described in the following.

The measured ratio of $A_{43}/A_{48}$ of natural abundance appeared to be 0.62 although its tabulated value is 0.72. We repeatedly measured the ratio and observed ±3% scattering on average. We take 0.62 as a standard value and our discussion here is limited to 3% in accuracy. The results are for a counter-current speed of 0.7 mm/s, which corresponds to the mobility at 120 V/cm. At 170 V, we obtained a peak value of 3.08 for $\alpha(48/43)$, which is a surprisingly large number. The voltage applied to the 1 cm thick BN plate is estimated to be ~80% of the total voltage between electrodes. It is thus 136 V/cm for the applied voltage of 170 V. The peak appears at the voltage a little higher than that expected from the speed of the counter-current flow.

Here, note that calcium ions in the extracted solution are only small fraction of the original solution. The dominant fraction goes downwards through the cation exchange membrane as waste. We thus expect almost no change in the enrichment of the waste. Actually, we observed no change within error.

4.2. Mass dependence

Here we measure the ratio of $A_{48}/A_{43}$. The enrichment factor that is useful in practice is the ratio of $\alpha(48/40) = A_{48}/A_{40}$. The enrichment factor is the result of multiplication of the separation coefficient for a unit migration length. The mass dependence of the separation coefficient is known to be linear [8], and thus the estimated enrichment factor $\alpha_{est}(48/40)$ is reasonably given by

$$\alpha_{est}(48/40) = \exp\left(\frac{8}{5} \ln(\alpha(48/43))\right). \quad (7)$$

This equation gives 6 for $\alpha_{est}(48/43)$. We would like to emphasize here that 6 for the enrichment factor of $^{48}$Ca over $^{40}$Ca is a big number for this kind of simple instrument.
Table 1. Values of abundance ratios $A_{43}/A_{48}$, enrichment factor of $\alpha_{\text{est}}(48/43)$ and $\alpha(48/43)$ are shown.

| applied voltage (V) | $A_{43}/A_{48}$ | $\alpha(48/43)$ | $\alpha_{\text{est}}(48/40)$ |
|--------------------|----------------|----------------|-----------------|
| 180                | 0.538          | 1.15           | 1.25            |
| 170                | 0.201          | 3.08           | 6.05            |
| 160                | 0.387          | 1.60           | 2.12            |
| 150                | 0.583          | 1.06           | 1.1             |
| natural            | 0.62           | 1.0            | 1.0             |

Fig. 2. Enrichment factors are shown as a function of applied voltage. Diamonds (blue) show the measured enrichment factor of $\alpha(48/43)$ and squares (red) show the calculated enrichment factor of $\alpha_{\text{est}}(48/40)$ from $\alpha(48/43)$.

4.3. Power density and temperature

When we observed the electric current of 0.2 A, it was a rough mean value and varied by about 20% during the experiment. The total power in the migration path in the BN plate is 27 W ($= 136 \times 0.2$). Since the volume of the migration path is 0.34 cc, the power density is 80 W/cc. This can be compared with CCE, where $\sim 0.1$ W/cc was applied to keep the temperature at around 80 degrees [7]. MCCCE achieved an increase of almost 3 orders of magnitude in the power density over CCE. The calculated temperature rise in each channel ($\Delta T_{\text{ch}}$) is 5 degrees and that in BN ($\Delta T_{\text{BN}}$) is 4 degrees. The total temperature rise is 9 degrees, which keeps the temperature safely lower than 100 °C anywhere. There is room to tolerate a higher power density. MCCCE achieved a 3 order-of-magnitude increase in the power density over CCE and also achieved a 3.5 order-of-magnitude increase in the cross-sectional area ($(0.8/0.1)^2 \times 69 = 4400$) over CE. A further increase is possible with a BN plate with higher $\lambda_{\text{BN}}$ and bigger cross-sectional area.

5. Discussion

5.1. Conditions

We need to mention that these remarkable results were obtained under subtly controlled conditions. We obtained the result as a function of the applied voltages. The voltage that showed maximum enrichment varied by about 10% and the enrichment appeared to be slightly different. Conceivable reasons for this are as follows. Firstly, bubbles were produced when we applied voltage. Most of them were produced near the electrodes. Bubbles at the cathode (bottom) were safely removed by the
cation exchange membrane and those at the anode (top) caused no harm in this experiment. However, bubbles produced at the lower surface of the BN plate blocked some fraction of the channels. They thus changed the flow speed and changed the balance between migration speed and flow speed. We tried to remove the bubbles by setting the instrument horizontally. Although the bubbles were successfully removed, we observed little enrichment factor. Convection of aqueous flow might mix the original solution (below the BN) and the enriched solution (above the BN). Although we do not completely understand the reason for this, we concluded that vertical setting of the instrument was important for the current instrument. We finally set the instrument vertically to obtain a good enrichment factor and tilted it slightly (10–15 degrees) to remove the bubbles. We were able to control the bubbles reasonably well but not completely. We think that this is a reason why we observed a good enrichment factor but the peak voltage appeared to be slightly different. We plan to achieve complete control of the bubbles in a new instrument.

5.2. Peristaltic pump

Secondly, our pump system does not have complete control of the flow speed. We used a peristaltic pump for the aqueous flow. The peristaltic pump makes a flow by a circular rotor pressing a flexible tube. The flow consists of pulses sent every 3–4 seconds. It is not smooth, accurate, or stable since the tube is sometimes deformed. The peristaltic pump could be another reason why the flow speed and voltage relation varies slightly.

Once we obtained remarkable results, we replaced the peristaltic pump with a plunger pump, which can give smooth, stable, and accurate flow. However, the remarkable results disappeared. It took a long time for us to recognize that the peristaltic pump was key to obtaining a good enrichment factor. This was due to the velocity distribution of the counter-current flow, as described in the following.

5.3. Hagen–Poiseuille flow

In order to obtain good separation, the speed of the flow should be uniform at any point in a channel or the flow front should be flat. However, the flow is laminar in the channel to satisfy the boundary condition that the speed is zero at the surface. This is the so-called Hagen–Poiseuille flow. For a cylindrical migration path of radius $a$, the speed has radius dependence, represented by

$$v(r) = 2v_0 \left(1 - \frac{r^2}{a^2}\right),$$

where $v_0$ is the average speed. Its standard deviation (width) is $\sigma_v = \frac{1}{2}v_0 = \frac{1}{\pi a^2} \int_0^a v(r)2\pi r dr$. The velocity (mobility) difference between isotopes, on the other hand, is of the order of %. Therefore, tiny differences in the migration lengths between isotopes would be wiped out by the Hagen–Poiseuille flow.

The condition of whether the flow becomes laminar or turbulent is determined by the Reynolds number $Re$, which is defined as

$$Re = \frac{vd}{v},$$

where $v$ is mean velocity, $d$ is diameter, and $v$ is the kinematic viscosity of water. In this experiment, $v$ is 0.7 mm/s, $d$ is 0.8 mm, and $v$ is $1 \times 10^{-6}$ [m$^2$/s]. They give $Re \sim 0.5$. Usually the flow is laminar when $Re$ is less than a couple of thousand. Therefore, the counter current in the present experimental condition is clearly laminar for the static flow. In principle, laminar flow is desirable since turbulent flow further mixes the migration length difference between isotopes. However, we need to have flow with a flat flow front.
We obtained a remarkable enrichment factor when we used a peristaltic pump but not a plunger pump. In other words, the pulsed flow generated by a peristaltic pump relaxes the boundary condition of the laminar flow and the flow becomes uniform. Use of a peristaltic pump effectively solved the problem of flatness of the flow front.

5.4. Oscillating flow in time and space

Laminar flow is unavoidable in MCCCE because of the Reynolds number, derived from the dimensions of the instrument and the counter-current speed. However, it is effectively solved by the pulsed flow, although its condition is subtle. When we speed up the pulse rate to, e.g., 1 pulse/s, we see little enrichment factor. Thus pulsed flow becomes continuous at a certain rate.

We consider that variation or oscillation of flow in both time and space help to make the flow front flat. We made the migration path on a 20 mm thick BN plate by piling up 2 mm thick BN plates of 1 mm\(\phi\) holes and 2.5 mm\(\phi\) holes alternatively. Brief optimization of the parameters gave 1.4 for \(\alpha(48/43)\). This means that oscillating flow in space has a similar effect to that of pulsed flow. However, the quantitative argument needs further study.

We made a 20 mm thick BN plate with 0.8 mm\(\phi\) holes. After brief optimization of the parameters, we obtained 4.2 for \(\alpha(48/43)\), which means 10 for \(\alpha(48/40)\). This is the enrichment factor that we originally set as the first-stage goal of CANDLES. Further details will be described in future publications.

Here we give a qualitative description of what we observed in our experiment; quantitative discussion of the flatness of the flow front is left for future study.

6. Summary

We developed the MCCCE method for the enrichment of large amounts of ions. The essential point of MCCCE is to increase power density and cross-sectional area by making multi-channel migration paths on an insulating material with high thermal conductivity. Here we used a BN plate that satisfies the requirement. We made a prototype instrument that realized the concept of MCCCE. Enrichment of \(^{48}\)Ca was carried out by the instrument, obtaining a remarkably high enrichment factor of 6 for \(\alpha_{\text{est}}(48/40)\). The tolerated power density was 80 W/cc, which is 3 orders of magnitude larger than that of CCE. The instrument also achieved a 3.5 order-of-magnitude increase in cross-sectional area over CE. The results from the prototype instrument demonstrate that MCCCE works as predicted. We found that the peristaltic pump was the key to the remarkable enrichment. Oscillating flow in either time or space makes the flow front flatter for the so-called Hagen–Poiseuille flow.

We need further development of the instrument and operation for the realistic enrichment of large amounts of ions. The bubbles need to be taken care of. Horizontal setting of the present instrument would still be seriously considered, since it is the simplest solution for the bubbles. The instrument has to be scaled up to process realistic amounts of ions.

It is obvious that one can apply this method to any other elements or compounds that can be electrically charged ions in aqueous solutions. If it is applied to nuclear fuel like uranium, one might make much smaller plant. It could also be applied to separation of radioactive isotopes on many occasions.

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References

[1] A. S. Barabash, Phys. Usp. 57, 482 (2014) and references therein.
[2] S. Umehara et al., EPJ Web Conf. 66, 08008 (2014).
[3] T. Kishimoto, Int. J. Mod. Phys. E 10, 2129 (2009).
[4] T. Kishimoto et al., Proc. 4th Int. Workshop on Neutrino Oscillations and their Origin, p. 338 (2003).
[5] A. K. Brewer, et al., J. Res. Nat. Bur. Stand. 38, 137 (1947).
[6] G. Kemp, Biotechnol. Appl. Biochem. 27, 9 (1998) and references therein.
[7] N. Kobayashi, Y. Fujii, and M. Okamoto, J. Chromatogr. 252, 121 (1982).
[8] Y. Fujii, J. Hoshi, H. Iwamoto, M. Okamoto, and H. Kakihana, Z. Naturforsch. A 40a, 843 (1985).