Isotope Effects in Electron Exchange Reactions of Lanthanide and Actinide Elements Including Chemical Uranium Enrichment*

YASUHIKO FUJI1, MASAO NOMURA1, TATSUYA SUZUKI1, DAMIAN AXENTE2*

1Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, O-okayama, Meguroku, Tokyo 152-8550 Japan
2National Institute for Research and Development of Isotopic and Molecular Technologies, 65-103 Donath Str., 400293, Cluj-Napoca, Romania

Isotope separation by chemical exchange is very popular for light elements, such as hydrogen, lithium, boron, carbon, nitrogen and oxygen. Due to the mass dependence of the isotope effects, the chemical exchange was considered not to be applicable to uranium enrichment. However, uranium enrichment processes by ion exchange and solvent extraction have been developed in Japan and France, respectively. The principle of the chemical uranium enrichment was not known for many years. It was clarified 10 years ago that the origin of the uranium isotope effects was not in the molecular vibration but in the electron-nucleus interaction. Since then, attention has been placed on the similarities between the isotope effects in chemical exchange reactions and the isotope shifts in atomic emission spectra. Study on the temperature dependence of the isotope separation factor was a key to separate the “electron effects” and “molecular effects”. In the present paper, experimental work on gadolinium isotope effects in amalgamation is presented and isotope effects of lanthanide elements and actinide elements are reviewed. Discussion is extended to the role and effects of “f” electrons of these elements. Non-proliferation is the most serious problem in nuclear technology development. The chemical exchange uranium enrichment has been regarded as the proliferation-resistant technologies. Chemical enrichment, whose development has been abandoned in Japan and France, could be reconsidered again among international nuclear technology communities.

Keywords: isotope effect, ion exchange, chemical exchange, uranium enrichment, non-proliferation, gadolinium, amalgam, oxidation-reduction reaction

Isotope effects have been drawing attention in different scientific fields. Recent research activities relating to isotope effects have been reviewed [1]. Isotope separation by chemical exchange is an application of isotope effects and very popular among light elements, such as hydrogen, lithium, boron, carbon, nitrogen and oxygen. For long time, the chemical exchange isotope separation was considered not to be applicable to uranium enrichment, due to the mass dependence of the isotope effects. However, uranium enrichment processes by ion exchange and solvent extraction have been developed in Japan and France, respectively, in the period of around 1970 – 1990 [2]. The principle of the chemical uranium enrichment was not known for many years. It was clarified 10 years ago that the origin of the uranium isotope effects was not in the molecular vibration, which was considered to be the key by the standard isotope effect theory, but in the electron-nucleus interaction, which was known as the source of isotope shifts in atomic emission spectral lines [3-6]. Since then, attention has been placed on the similarities between the isotope effects in chemical exchange reactions and the isotope shifts in atomic emission spectra. Because the nuclear-electron interaction is enhanced in the large and deformed nucleus, the chemical isotope effects of lanthanide elements are interesting topics. In the present paper, experimental work on gadolinium isotope effects in amalgamation are presented and isotope effects of lanthanide elements and actinide elements are reviewed. Non-proliferation of nuclear weapons is the most serious problem in nuclear technology development.

The chemical exchange uranium enrichment has been regarded as the proliferation-resistant technologies. Chemical enrichment may be again reconsidered by international nuclear technology communities in the future. In the present work the non-proliferation aspects are discussed on ion exchange uranium enrichment process.

Uranium isotope enrichment by chemical exchange
Uranium enrichment by chemical exchange has been studied on several processes. There are two most extensively studied processes: one is the solvent extraction process based on the isotope effects in U(III)-U(IV) exchange reaction and the other is ion exchange process, based on the isotope effects in U(IV)-U(VI) exchange reaction. The isotopic exchange reactions of these electron exchange systems are expressed by relations (1) and (2):

\[ ^{235}\text{U}(\text{III}) + ^{238}\text{U}(\text{IV}) \leftrightarrow ^{238}\text{U}(\text{III}) + ^{235}\text{U}(\text{IV}) \tag{1} \]
\[ ^{235}\text{U}(\text{IV}) + ^{238}\text{U}(\text{VI}) \leftrightarrow ^{238}\text{U}(\text{IV}) + ^{235}\text{U}(\text{VI}) \tag{2} \]

Isotope effects in U(IV) and U(VI) complex formation have been also intensively studied. In the reactions (3) and (4), complex species is expressed by ligand L:

\[ ^{235}\text{U}(\text{IV}) + ^{238}\text{U}(\text{IV}) L \leftrightarrow ^{238}\text{U}(\text{IV}) + ^{235}\text{U}(\text{IV}) L \tag{3} \]
\[ ^{235}\text{U}(\text{VI}) + ^{238}\text{U}(\text{VI}) L \leftrightarrow ^{238}\text{U}(\text{VI}) + ^{235}\text{U}(\text{VI}) L \tag{4} \]
Their isotope effects, or isotopic fractionation factors of these reactions, are quite small compared with those of electron exchange reactions and the research work was limited to the study on basic science.

Isotope separation based on U(IV) – U(VI) exchange system had been initially studied by using cation exchange resins in several countries and later industrial development work was conducted by using special anion exchange resin. Chromatographic uranium enrichment system is theoretically possible by using both cation and anion exchange resins. The reason, why the anion exchange resin was finally selected, was mainly due to the ion exchange selectivities of ionic species involved in the systems. In the cation-exchange resin method, a sharp boundary is clearly developed between U(VI) and Ti(III) boundary, where $^{235}$U is enriched, a sharp boundary is difficult to be maintained. The cation-exchange system is easy to produce depleted uranium at the U(IV)-Fe(III) boundary, but difficult to produce enriched uranium at the U(VI)-Ti(III) boundary. In the anion-exchange resin system, on the other hand, a sharp boundary is maintained between the uranium adsorption band and the reducing reagent zone. In this respect, anion-exchange resin system is appropriate to produce enriched uranium.

**Isotope effects of lanthanide elements**

The research into isotope separation of uranium by ion exchange led to the findings of the anomalous isotope effects, or mass dependence anomaly in isotope effects [3,4]. This phenomenon is called as “odd–even staggering”, which is well known in the isotope shift of atomic emission spectra. Since this phenomenon relates to the volume and shape of nuclei, results of the study on uranium isotope effects stimulated the isotope effects of other heavy elements and lanthanide elements. Lanthanide elements resemble actinide elements in the electron configuration, and gadolinium isotope separation by laser was studied as the simulation of uranium isotope separation because of its oxidation characteristics.

Gadolinium isotope effects in complex formation are studied by using cation-exchange resin. The odd isotopes $^{155}$Gd and $^{157}$Gd showed a slight deviation from the linear relation observed for even isotopes of $^{154}$Gd, $^{156}$Gd and $^{158}$Gd, [7]. The pattern of the deviation closely resembles the nuclear volume deviation expressed with the mean square nuclear radius. It has been concluded that the field shift isotope effects, which were discovered in the study on uranium isotope effects, are also predominant in isotope effects in gadolinium complex formation. Then attention has been placed on the amalagamation formation. The amalgamation formation has been known to show large isotope effects. Enriched lithium isotopes have been produced in an industrial scale by amalgam electrolysis. Light isotopes are enriched in amalgam phase, while the heavier isotopes are enriched in the solution phase among light elements such as Mg, K, Rb, [8-10]. On the other hand, the heavier isotopes are enriched in the amalgam phase in the case of barium, [11]. It is interesting to know the isotope effects in amalgamation formation of lanthanides, in particular, gadolinium.

**Experimental part**

**Uranium**

In the isotopic fractionation on the anion-exchange resin $U$(VI) is in the form of chloro-complexes of $\text{UO}_2^{2+}$, minus ions, and adsorbed in the anionite, while $U$(IV) is in the form of $\text{U}^{4+}$ aqua-chloro-complex, plus ion, and stays in aqueous phase. Either in process of $U$(IV)-$U$(VI) or in $U$(III)-$U$(IV) exchange system, hydrochloric acid is used as the working acid reagent in the aqueous solution. This is due to the appropriate oxidation-reduction condition of the acid. Nitric acid, which is used in reprocessing of spent fuel, is not usable in chemical uranium enrichment systems, because of its oxidation characteristics.

The unit stage separation factor of $U$(IV)-$U$(VI) exchange system being very small, the separation factor was multiplied by using displacement type chromatography. The anion exchange resin packed in a column is initially charged with Fe(III) and then U(VI) is fed into the column, until sufficient length of the uranium adsorption band is formed. Thereafter an eluent, containing reducing reagent, V(III), was fed into the column to elute the uranium adsorbed in the resin; adsorbed U(VI) is reduced to U(IV) and eluted to the aqueous phase. The reaction between U(VI) and V(III) is expressed by the reaction (5):

$$ U(\text{VI}) + V(\text{III}) \rightarrow U(\text{IV}) + V(\text{IV}) \quad (5) $$

The reduced species U(IV) is released from the resin phase. Both U(IV) and V(IV) are in the solution phase and flows down through the uranium adsorption band.

In the zone of the uranium adsorption band, the isotope exchange reaction takes place between the uranium species in the moving phase of the solution and the uranium species in the anion exchange resin. The isotopic reaction is expressed by the exchange equilibrium (6):

$$ 238\text{U}_{(\text{sol})} + 235\text{U}_{(\text{R})} \leftrightarrow 238\text{U}_{(\text{sol})} + 235\text{U}_{(\text{R})} \quad (6) $$

The uranium species expressed by $U_{(\text{sol})}$ represent uranium ions in the solution phase and composed of U(IV), as a major species and U(VI), as minor species. In the same way, the species represented by $U_{(\text{R})}$ are uranium ions in resin phase and composed of $U(\text{VII})$, as a major species and U(IV), as a minor species. In the ideal case where both phases contain only major species, and therefore the minor species do not exist, reaction (6) is consistent with reaction (2). The equilibrium constant of reaction (2) gives ideal isotope separation factor $\alpha$ between U(IV) and U(VI), while reaction (6) gives a practical separation factor $\alpha$ of a unit stage of the ion exchange system.

Uranium species U(VI) in the solution reaches the front boundary between the uranium adsorption band and Fe(III) adsorption band. The following oxidation reduction reaction takes place and oxidized uranium, U(VI), is adsorbed in the resin:

$$ U(\text{IV}) + \text{Fe(III)} \rightarrow U(\text{VI}) + \text{Fe(II)} \quad (7) $$

Generated Fe(II) is released to the solution phase, and flows down through the Fe(III) adsorption band. The solution contains not only Fe(II) but also V(IV), which is generated by the reduction of U(VI). At the front of the Fe(III) adsorption band the following reaction takes place:

$$ V(\text{IV}) + \text{Fe(II)} \rightarrow V(\text{III}) + \text{Fe(III)} \quad (8) $$

Although Fe(II) is not completely oxidized to Fe(III), reaction (8) suggests that the reducing reagent V(III)
and the oxidizing reagent Fe(III) can be partly regenerated. These reactions are summarized in figure 1.

Practical enrichment of uranium isotopes by ion exchange was attained by improvement in kinetic factors of ion exchange reaction rates and electron exchange reaction rates. Each phase of the ion exchange resin and the aqueous solution contains one of U(IV) and U(VI) as major species and the other species as minor component. Isotopic ion exchange reaction takes place between U(VI) in resin phase and U(VI) in solution phase. In the same way, isotopic ion exchange reaction takes place between U(IV) in solution phase and U(IV) in resin phase. In each phase, U(IV) and U(VI) exchange their electrons. The electron exchange reaction is expressed by reaction (2), when isotopes are concerned. Via electron exchange reactions and ion exchange reactions, uranium in resin phase and uranium in solution phase are exchanged and the isotopic equilibrium of reaction (6) is attained. These exchange relations are summarized in figure 2.

Uranium enrichment chromatography was conducted at 160°C in an experimental plant with four titanium alloy columns (2cm I.D., 107 cm resin bed height each) with oil jackets for temperature controlling [6]. Commercially available anion exchange resins are not usable at this high temperature. The porous small particle (ca. 60 μm), benzimidazole quaternary ammonium ion type resin was specially developed by Asahi Chemical Industries and used in present work. In table 1 are listed the experimental conditions of chromatographic operation at 160°C.

Results and discussion

Gadolinium

Experimental work was conducted on the isotope effects in gadolinium amalgam electrolysis. The formation of gadolinium amalgam was experimentally studied [12]. The present work follows their experimental conditions of gadolinium amalgam electrolysis. Gadolinium metal was dissolved in acid, and converted to gadolinium acetate. Lithium citrate was added to the electrolyte solution containing gadolinium acetate at the ratio of [Li] / [Gd] = 12. The electrolysis cell, shown in figura 3, is made of glass with a water jacket. The inner diameter of the cell was 40 mm and the current density of mercury electrode (cathode) was ca. 0.032 A/cm². Platinum wire was used as the anode. Temperature was controlled at 25°C by thermostated water, circulated through the water jacket.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>EXPERIMENTAL CONDITIONS OF CHROMATOGRAPHIC ENRICHMENT OF 235U</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>160</td>
</tr>
<tr>
<td>Migration distance (cm)</td>
<td>428</td>
</tr>
<tr>
<td>Eluent feed rate (cm³/min)</td>
<td>7.9</td>
</tr>
<tr>
<td>Band velocity (m/h)</td>
<td>0.90</td>
</tr>
<tr>
<td>Oxidizing solution (cm³)</td>
<td>300</td>
</tr>
<tr>
<td>Uranium feed solution (cm³)</td>
<td>350</td>
</tr>
<tr>
<td>Reducing eluent (cm³)</td>
<td>2,300</td>
</tr>
</tbody>
</table>

Uranium

The chromatographic profile and the measured isotopic abundance ratios of 235U/238U in the uranium migration band are shown in figure 4 for experimental run at 160°C. The lighter isotope 235U is enriched at the rear boundary, which means 235U is preferentially fractionated in U(IV) or UO₂²⁻ ion, while the heavier 238U is enriched in the front boundary. Based on the experimental results, the separation coefficient ε was calculated for 160°C as 0.0008, with an error estimated as 15%. It was determined that the isotopic enrichment of 235U is improved with increase in the operational temperature. The higher temperatures are efficient conditions for production of enriched uranium by the present process. The separation coefficient ε is important from the
In Run 1, the electrolysis solution was made by mixing 7.5 ml of 0.02M Gd(Ac)$_3$, 4.5 ml of 0.4 M Li$_3$Cit and 3 ml of H$_2$O. The volume of mercury was 10 mL. The electrolysis experiment was conducted at the applied voltage, 13-15 V and at the current 0.25- 0.43 A. After the electrolysis, the mercury phase was taken and stirred with 1 M HCl. Then the gadolinium solution was precipitated as oxalate. Finally gadolinium was converted to Gd$_2$O$_3$. The gadolinium oxide sample 0.01 g was obtained and subjected to mass analysis. The remaining gadolinium in the solution phase was also precipitated as oxalate and converted to Gd$_2$O$_3$ of which amount was 0.0059 g. The electrolysis yield of gadolinium amalgam was 37 %. The experimental conditions of Run 2 are basically the same as those of Run 1. The electrolysis time was expanded to 230 min. In Run 3, the same amounts of Gd solution and Li solution was used, but added water is increased to 28 ml. Amount of mercury was also increased to 20 ml. Since the concentration of aqueous solution was decreased, slightly higher voltage of 14 – 17.5 V was applied in the electrolysis. In Run 4, both amounts of Gd and Li solutions were reduced to a half of the initial conditions in Run 1 and 14 mL of H$_2$O was added. The concentration of Gd was, therefore, further reduced and the electrolysis was carried out at the applied voltage of 25 – 34 V.

The samples of gadolinium oxide recovered after the electrolysis experiments were subjected to mass spectrometric analysis for measurement of isotopic abundance ratios. The results of the isotopic analysis for isotopic pair $^{156}$Gd and $^{160}$Gd are listed in table 2 along with the recovered amount of Gd$_2$O$_3$ from each phase. As the average of four runs, the isotopic separation factor, or isotopic fractionation factor between aqueous phase and amalgam phase, $\alpha$ is determined as 1.0024. The observed isotopic separation factor clearly shows that the light isotope is enriched in the amalgam phase.

**Isotope effects and electron configuration**

In a previous work on Eu(II)-Eu(III) isotope exchange experiments, light isotope was enriched in higher valence state of Eu (III), [13]. This tendency according to U(III) – U(IV) and U(VI) – U(VI) exchange systems. These exchange systems indicate light isotopes are enriched in higher valence states, which have larger number of f electrons. The isotopic distribution of the present experimental results is opposite; lighter isotope is enriched in lower valence state of Gd metal (alloy). In the Gd(0) – Gd(III) exchange system, the number of f electron is constant and Gd(0) has more 5d and 6s electrons in the outer shell while Gd(III) does not have 5d and 6s electrons. The experimental results, showing that light isotope is preferentially enriched in Gd(0) state, can be explained by the field shift isotope effect, which is originated from the presence of s electrons at the nucleus.

So far isotope effects have been observed in several electron exchange systems of lanthanide and actinide.

---

### Table 2

<table>
<thead>
<tr>
<th>Run</th>
<th>Recovered Gd$_2$O$_3$(mg)</th>
<th>Isotopic ratio $^{156}$Gd/$^{160}$Gd</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Amalgam 10</td>
<td>0.9485</td>
<td>1.0023</td>
</tr>
<tr>
<td></td>
<td>Water</td>
<td>0.9463</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Amalgam 7.6</td>
<td>0.9494</td>
<td>1.0013</td>
</tr>
<tr>
<td></td>
<td>Water</td>
<td>0.9482</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Amalgam 13.1</td>
<td>0.9468</td>
<td>1.00456</td>
</tr>
<tr>
<td></td>
<td>Water</td>
<td>0.9425</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Amalgam 11.4</td>
<td>0.9484</td>
<td>1.0013</td>
</tr>
<tr>
<td></td>
<td>Water</td>
<td>0.9472</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td>1.0024</td>
</tr>
</tbody>
</table>

---

**Number of electrons**

<table>
<thead>
<tr>
<th>$f$</th>
<th>$s$</th>
<th>Constant</th>
<th>Increase</th>
</tr>
</thead>
</table>

**Chemical System**

- U(II) – U(III)
- Gd(II) – Gd(III)
- Yb(II) – Yb(III)
- Eu(II) – Eu(III)

**Isotope Fractionation**

- Light → Heavy
- Heavy → Light
It is interesting that the reported results commonly indicate that light isotope is enriched in the species, which has larger number of s electrons or smaller number of f electrons. The reason, why the lighter isotope is enriched in the s electron rich side, is easily explained by the field shift effect theory. On the other hand the reason, why the lighter isotope is enriched in the f-electron depleted side is complicated. Probably f electrons shield the positive charge of nucleus and reduce the probability of outer s electrons at the nucleus. The isotopic fractionations and electron configurations are summarized in table 3. The problems of the role of f electrons are expected to be solved in the future by theoretical study on the isotope effects of heavy elements.

Proliferation resistance characteristics

It should be noted that the ion exchange uranium enrichment has excellent characteristics from a viewpoint of non-proliferation policy, [14]. Firstly, the product enrichment is limited by the criticality of the system. The commercial plant is anticipated to be operated with large diameter columns packed with anion exchange resin. Criticality considerations on 1 m diameter column are shown in figure 5. In the case of anion-exchange resin system, Case II in figure 5, the maximum enrichment degree of the product is approximately 50%, since the neutron multiplication factor should be maintained at less than 0.9 from the safety reason while in operation.

Secondly, in addition, production of highly enriched uranium is time consuming. It is due to the small separation factor and the large amount of uranium inventory necessitated at the enriched part in the column. The plant equilibrium time or the duration of time for production of enriched uranium from the start of the enrichment work using natural uranium is calculated assuming the parameter values for operation and presented in figure 6, which includes hypothetical cases of large separation coefficient and the high-speed migration, represented a and b in figure 6. The practical cases, c and d in figure 6, indicate that only 50% enriched uranium can be produced after ten years from the beginning of operation with natural uranium. To obtain 90% enriched uranium, the operation time necessitates 30 years or so. This means that the ion exchange process cannot produce highly enriched uranium.

Conclusions

By chromatographic uranium isotope enrichment experiment, conducted at 160°C, the separation coefficient ε was determined; it is consistent with data reported in previous work, [15] for the temperature range 50 – 87°C.

The isotopic enrichment of 235U was improved with increase in the operational temperature according to the higher isotope exchange rate at 160°C in the U(VI) – U(IV) system on special anion exchange resin.

The results of the isotopic analysis for isotopic pair 156Gd and 160Gd showed that the isotopic fractionation factor between aqueous phase and amalgam phase, α, is 1.0024, the light isotope being enriched in the amalgam phase.

From the view point of non-proliferation, the characteristics of ion exchange enrichment of 235U is summarized as:

- practically enrichment is limited to 50% 235U from the view point of the fission criticality;
- to produce highly enriched uranium by the commercial plant needs very long time, 10 years for production of 50% 235U enriched uranium. To produce 90% 235U, the necessitated plant operation time is increased to 30 years or so.
- IAEA safeguards is easy to be applicable since the facility is basically a chemical plant where the pipes are fixed and easy to be checked.

References


Fig. 5. Criticality of ion exchange uranium enrichment plant

Fig. 6. The equilibrium time of ion exchange uranium enrichment process
In perioada 17 – 21 august 2008 a avut loc la Philadelphia, Philadelphia Convention Center, cea de a 236-a întrunire națională a Societății Americane de Chimie. Cu ocazia acestei întruniri a fost organizată și o mare expoziție de aparatură din domeniul chimiei.

La întrunire au fost prezentate mai mult de 8.000 de lucrări în sesiuni orale programate pe 717 jumătăți de zi. Au fost de asemenea 93 de sesiuni pentru postere. În plus au fost numeroase sesiuni comune, recepții și simpozioane. Lucrările conferinței s-au desfășurat în aproximativ 10 locuri speciale din Philadelphia, cel mai important loc fiind Philadelphia Convention Center, amenajat în parte în cadrul fostei gări centrale, desfășinată datorită numărului redus de călători.


Sesiuni de comunicări au fost organizate de asemenea de următoarele secțiuni și comitete ale American Chemical Society: Division of Agricultural & Food Chemistry, Division of Agrochemicals, Division of Analytical Chemistry, Division of Biochemical Technology, Division of Biological Chemistry, Division of Business Development & Management, Division of Carbohydrate Chemistry, Division of Chemical Education, Division of Chemical Health & Safety, Division of Chemical Technicians, Division of Chemical Toxicology, Division of Chemistry & the Law, Division of Colloidal & Surface Chemistry, Division of Computers in Chemistry, Division of Environmental Chemistry, Division of Fuel Chemistry, Division of Geochemistry, Division of the History of Chemistry, Division of Industrial & Engineering Chemistry, Division of Inorganic Chemistry, Division of Medicinal Chemistry, Division of Nuclear Chemistry & Technology, Division of Organic Chemistry, Division of Petroleum Chemistry, Division of Physical Chemistry, Division of Polymer Chemistry, Division of Polymeric Materials: Science and Engineering, Division of Professional Relations, Division of Small Chemical Businesses, Committee on Chemists with Disabilities, Committee on Corporation Associates, Committee on Environmental Improvement, Committee on Economic & Professional Affairs, Committee on Local Section Activities, Committee on Minority Affairs, Committee on Project SEED, Committee on Professional Training, Committee on Science, Society Committee on Education, Women Chemists Committee, Younger Chemists Committee.

Sunt de menționat și simpozioanele organizate de președintele American Chemical Society, Bruce E. Bursten dintre care menționăm: Global Energy Challenges in the 21th Century și Project SEED: Cutting-edge Research with High School Students.

În cadrul întrunirii de la Philadelphia au fost organizate de asemenea numeroase cursuri scurte pentru educația continuă a specialiștilor, workshop-urile profesionale și workshop-urile în cadrul expoziției. La expozitia organizată au participat peste 300 de firme.

A fost de remarcat calitatea deosebită a multora dintre comunicările prezentate la întrunire, cele mai multe dintre acestea abordând teme de mare actualitate ale chimiei contemporane.

La conferință au participat peste 8000 de specialiști în primul rând din SUA, dar și din Canada, Europa de Vest și Japonia. Numărul participanților din alte țări (Europa de Est, Rusia, India, China, America Centrală și de Sud etc.) a fost însă redus.

Prof. dr. Andrei Florin Dâneț
Universitatea din București
Facultatea de Chimie