Chemical consequences of the β -decay of ¹⁴⁷Nd in tris(cyclopentadienyl)neodymium

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Dedicated to the 70th birthday of Professor M. Dillinger

The main chemical form of the stabilization of ¹⁴⁷Pm arisen by the β -decay of ¹⁴⁷Nd in tris(cyclopentadienyl)neodymium was determined. The accumulation of daughter products took place in the crystals of parent compound and their separation was carried out by sublimation method. Under the conditions employed the main chemical form of the stabilization of ¹⁴⁷Pm is (C₅H₅)₃Pm which is formed in the yield of 91.3 \pm 3.5%. The remainder consists of some chemical forms which are not volatile at 180°C and 10⁻⁴ torr and have not been exactly defined as yet.

The study of the chemical processes accompanying a nuclear transformation of the atoms which represent a part of polyatomic molecules becomes an important field of radiochemistry. The primary products of a β -decay in neutral molecules are, at least in the first moment, molecular ions with a selectively localized charge. Later on, their destiny will especially depend on the value of recoil energy and probability of a subsequent internal ionization of a daughter atom, on the chemical properties of this atom and the physicochemical conditions under which this transformation takes place. The overall effect of these factors in an individual case may result in a total disruption of the chemical bonds "inherited" by daughter atom from parent molecule and thus in the stabilization of daughter atom in a chemical form which is quite different from that of parent atoms. However, these bonds are frequently fully or partially preserved (regenerated) and the daughter atom is stabilized in a chemical form which is analogous or near to the chemical form of parent atoms.

In several studies the chemical changes caused by β -decay were utilized as a means for the preparation and investigation of the properties of new compounds, in particular, the organic compounds of polonium [1-6], organometallic compounds of some artificial radioactive elements, namely technetium [7, 8], promethium [9], and neptunium [10].

The parent radionuclide may be obtained in a certain chemical form not only by classical chemical synthesis but also for instance by the neutron activation of stable isotopes of this element bonded in such chemical form in which the parent radionuclide should be bound. The neutron activation of natural samples of neodymium, however, affords three β -radioactive isotopes of neodymium which are the first members of three disintegration chains:

$$\frac{\beta^{-}}{T = 11.1 \text{ days}} \xrightarrow{147} \text{Pm} \xrightarrow{\beta^{-}}{T = 2.65 \text{ years}} \xrightarrow{147} \text{Sm} \xrightarrow{\alpha} \xrightarrow{143} \text{Nd (stable)},$$

$$\frac{149}{T = 1.2 \times 10^{11} \text{ years}} \xrightarrow{149} \text{Nd} \xrightarrow{\beta^{-}}{T = 1.8 \text{ hours}} \xrightarrow{149} \text{Pm} \xrightarrow{\beta^{-}}{T = 54 \text{ hours}} \xrightarrow{149} \text{Sm (stable)},$$

$$\frac{151}{T = 12 \text{ minutes}} \xrightarrow{151} \text{Pm} \xrightarrow{\beta^{-}}{T = 27.5 \text{ hours}} \xrightarrow{151} \text{Sm} \xrightarrow{\beta^{-}}{T = 93 \text{ years}} \xrightarrow{151} \text{Eu (stable)}.$$

The total activity of a temporarily activated sample is practically given at the end of activation only by the activity of the first two members of each chain. Provided the sample was activated for the period of time t, its activity $A_{t'}$ in arbitrary moment $t' \ge t$ is given by the expression

$$egin{aligned} &A_{t'} = \sum A_{ extsf{s} \ extsf{Nd}-i} [1 - \exp(-\lambda_{ extsf{Nd}-i} t)] \exp(-\lambda_{ extsf{Nd}-i} \, \Delta t) + \ &+ \sum A_{ extsf{s} \ extsf{Nd}-i} [1 - \exp(-\lambda_{ extsf{Pm}-i} t)] \exp(-\lambda_{ extsf{Pm}-i} \, \Delta t) + \end{aligned}$$

$$+ \sum A_{s Nd-i} \frac{\lambda_{Pm-i}}{\lambda_{Nd-i} - \lambda_{Pm-i}} \left[\exp(-\lambda_{Nd-i} t) - \exp(-\lambda_{Pm-i} t) \right] \exp(-\lambda_{Pm-i} \Delta t) +$$

$$+ \Sigma A_{s Nd-i} \frac{\lambda_{Pm-i}}{\lambda_{Pm-i} - \lambda_{Nd-i}} [1 - \exp(-\lambda_{Nd-i} t)] [\exp(-\lambda_{Nd-i} \Delta t) - \exp(-\lambda_{Pm-i} \Delta t)],$$

where A_s , *i*, λ , and Δt are saturation activity, mass number (equal to 147, 149, and 151), decay constant, and difference between t' and t, respectively.

At the end of a brief activation used by *Baumgärtner et al.* [9] ¹⁴⁹Nd and ¹⁵¹Nd take the main part in the total activity. But the relatively short half life of the decay of these radionuclides does not allow to differentiate by sublimation method the chemical changes in $(C_5H_5)_3$ Nd caused by (n, γ) reaction from the chemical changes induced by β -decay. Therefore the authors focused their attention only on the proof of the existence of $(C_5H_5)_3^{151}$ Pm without estimating the yield.

In order to investigate the chemical consequences of the β -decay of the central atom in $(C_5H_5)_3Nd$, we chose in this study the first of the above-mentioned chains which enabled us to eliminate the effect of (n, γ) reaction. The chemical effects caused by the β -decay of ¹⁴⁷Nd could be evaluated after a sufficiently long period of time since the end of neutron activation on the basis of activity measurement of the only radionuclide - ¹⁴⁷Pm.

Experimental

Chemicals

Dicyclopentadiene (Urx Works, Ostrava) was decomposed by heating into vapourof monomer, which condensed in a condenser after passing through a dephlegmator. The condensate was collected in an ice-cooled vessel.

Tetrahydrofuran (THF) anal. grade (Carlo Erba, Milan) was purified according to the method described in [11].

Neodymium(III) chloride, anhydrous, 99.9% (Koch-Light Lab. Ltd., England).

Molecular sieve Calsit 5 (Research Institute for Petroleum and Hydrocarbon Gases,

Bratislava) was activated by heating for 4 hours at 350°C before use and afterwards cooled *in vacuo*.

Argon 3N5 (Moravian Chemical Works, Ostrava) with the content of oxygen below 10 p.p.m. was dried by going through a batery of the columns filled with silica gel and molecular sieve.

All other chemicals used were anal. grade.

Apparatus

The mass-spectrometric analysis was performed with a mass spectrometer of the type MCH-1303 (USSR).

The β -activity of samples was measured with a scintillation counter VA-S-971 equipped with a plastic scintillator SPD-23 in connection with a VA-M-160 (VEB Vakutronik, GDR) counter.

 γ -Ray spectra were recorded by means of a one-channel scintillation γ -ray spectrometer VA-M-120 connected with a scintillation counter VA-S-963 (VEB Vakutronik, GDR) and a "well-type" NaI(Tl) scintillator SKW-1S.

Syntheses

All reactions and operations involving hygroscopic substances or the substances sensitive to air were carried out in a dry medium and argon atmosphere.

Sodium cyclopentadienide — CpNa was prepared in the form of pale-pink-coloured solution by using the reaction of dispersed metallic sodium (0.782 mole) with freshly distilled cyclopentadiene (0.811 mole) in THF [12]. The concentration of CpNa in THF was determined by the titration of NaOH liberated in 50% ethyl alcohol from CpNa by the effect of hydrolysis with 0.1 n-HCl. THF and ethyl alcohol were evaporated before the titration.

Tris(cyclopentadienyl) neodymium was prepared in small amounts (grams) by the reaction of anhydrous NdCl₃ and CpNa (molar ratio 1 5) in the medium of THF [13]. It was separated from the reaction mixture and purified by threefold sublimation at 180°C

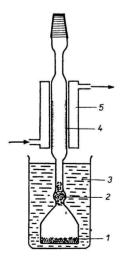


Fig. 1. Sublimation-reaction vessel for the preparation of labelled cyclopentadienyl derivatives.

1. reaction product; 2. glass wool; 3. silicone oil; 4. sublimate; 5. cooler. and 10^{-4} torr. For the synthesis of Cp₃Nd from NdCl₃ which was irradiated with neutrons, 100-150 mg of chloride were used and the whole preparation cycle of the pure product was carried out in the sublimation-reaction vessels presented in Fig. 1 which, when necessary, enabled us to preserve the obtained product for a longer time by sealing the upper and lower narrowed part of the vessel.

Neutron activation

The samples of NdCl₃ in the amount of 150–250 mg or Cp₃Nd in the amount of 60–150 mg sealed in an argon atmosphere into quartz ampoules were activated for 20 hours in a nuclear reactor of the type VVR-S (Institute of Nuclear Research of the Czechoslovak Academy of Sciences, Řež near Prague) at a neutron flux of about 10¹⁶ m⁻² s⁻¹. The temperature in irradiation channel was about 80°C and the integral dose of γ -radiation was approximately 10⁸ rad.

Analysis of products

The qualitative analysis of Cp_3Nd was performed by measuring the mass spectra. For this reason, the samples weighing 20-30 mg were inserted into the feeding system of mass spectrometer in evacuated sealed glass ampoules which were heated to working temperature and broken. The spectrum was measured at the ionization voltage of 30 V at 300 °C.

After the decomposition of the weighed samples in a hot mixture of HNO_3 and H_2O_2 , neodymium was quantitatively determined by the precipitation in the form of oxalate which was ignited under formation of oxide. The presence of chloride was determined by means of silver nitrate.

Radiometric measurements

The radioactive purity of samples was tested by measuring the γ -ray spectra as well as the absorption and decay curves obtained by detection of β -radiation. The measurements of the γ -ray spectra were carried out with the samples of Cp₃Nd which was obtained either by the synthesis from NdCl₃ activated with neutrons or the sublimation of the ready-made Cp₃Nd activated with neutrons. The measurements with the same samples were several times repeated in fixed time intervals. The samples for the measurement of β -radiation were obtained by dissolving the weighed samples of active Cp₃Nd prepared by one of the above methods in a mixture of concentrated HNO_3 and H_2O_2 . After filling up with water to a certain volume such amounts of solution were put on aluminium plates that the thickness of solids after evaporation did not exceed 0.002 - 0.005 kg m⁻². Using β -radiation the radioactive purity of ¹⁴⁷Nd was tested by measuring the β -decay curves of preparations after 20 days since the activation had been finished. At first, the counting rate of the preparation was measured without a filter and subsequently with an aluminium absorber (thickness 0.55 kg m^{-2}) which absorbed totally the β -radiation of ¹⁴⁷Pm. The radioactive purity of ¹⁴⁷Pm in the preparations of Cp₃Nd was tested before as well as after sublimation by measuring the absorption curves of the collimated β -radiation of 147 Pm by means of aluminium absorbers the thickness of which was 0.026 kg m⁻². These measurements were, however, performed after the β -decay of ¹⁴⁷Nd was completed.

A certain part of Cp_3Nd was taken immediately after its activation, that is to say, before as well as after sublimation with the view of determining the practical yield of ¹⁴⁷Nd in the form of Cp_3Nd .

Separation of the chemical forms of ¹⁴⁷Pm

The samples of Cp₃Nd were put into the medium part of a sublimation-reaction vessel which was sealed on both sides (Fig. 1). Provided Cp₃Nd was prepared from NdCl₃ activated with neutrons, this operation was preceded by a threefold sublimation through a layer of glass-wool whereas a single sublimation was applied in the case of Cp₃Nd which was activated with neutrons. Then the ampoules were allowed to stand for 140–200 days in the course of which a full β -decay of ¹⁴⁷Nd was achieved. Afterwards the ampoules were opened. A certain part of Cp₃Nd was taken for radiometric analysis and the remainder was sublimed again. The sublimate was used for the preparation of new samples for radiometric analysis.

Results and discussion

The preparation method by which Cp₃Nd was obtained for the first time [13] en abled us to obtain the needed product in good yield as soon as after 2-3 hours' reaction time. This was important especially in that series of experiments for which Cp₃Nd was synthesized from NdCl₃ activated with neutrons. After a threefold sublimation the preparations of Cp₃Nd containing $41.9 \pm 0.5\%$ Nd (calculated 42.5% Nd) were obtained (m.p.

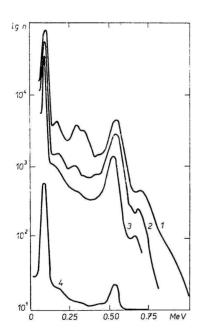


Fig. 2. γ -Ray spectra of $(C_5H_5)_3Nd$ activated with neutrons.

1. 6 days after activation; 2. 13 days after activation; 3. 22 days after activation;
4. 97 days after activation.

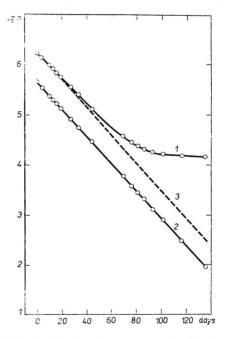


Fig. 3. Disintegration curves obtained by measuring β -radiation of $(C_5H_5)_3Nd$ activated with neutrons.

1. without absorber; 2. with an aluminium absorber of 0.55 kg m⁻² thickness; 3. extrapolated disintegration curve of ¹⁴⁷N⁻ without absorber. $382 + 1^{\circ}$ C, according to literature 380° C). The presence of chloride anion was not proved in these preparations. The mass-spectrometric analysis in the region of heavier fragments (containing the atom of neodymium) showed only the presence of the ions arising from Cp_3Nd while the fragmentation involved in preference by disruption of the Cp-Ndbonds. The relative intensities of main ions were as follows: $[Cp_3Nd]^+ = 21.9\%$, $[Cp_2Nd]^+ = 100\%$, $[CpNd]^+ = 44.4\%$ and $[Nd]^+ = 2.2\%$. These data conform with the analogous data on praseodymium and neodymium [14, 15]. According to some authors [16] it is possible to draw conclusion on the ionic character of the chemical bond between a metal atom and organic ligand on the basis of the character of mass spectrum of organometallic compounds. From this point of view the data obtained from the mass spectrum of Cp₃Nd are in agreement with the conclusion that tris(cyclopentadienyl)lanthanoides are compounds of ionic character [13]. The authors came to this conclusion on the basis of magnetochemical and spectrophotometrical measurements, some chemical reactions, and high melting points of these substances. They are very sensitive to air and humidity and are either instable or almost insoluble in most common organic solvents. Later we demonstrated in the case of Cp₃Nd [17] and Cp₃Pr [18] that a rapid homogeneous isotopic or ionic exchange of central atom might take place in some solvents. For this reason the sublimation method was chosen as a convenient and simple separation method which might distort very little the primary state of the assumed chemical forms of ¹⁴⁷Pm stabilization. Cp₃Nd labelled with ¹⁴⁷Nd was obtained either by a rapid synthesis from freshly activated $NdCl_3$ or by the sublimation of activated Cp_3Nd . In the γ -ray spectra of Cp₃Nd samples (Fig. 2) we found only the maxima corresponding to the γ -transitions accompanying the β -decay of corresponding radionuclides of neodymium and promethium. After a period of time exceeding the thirteenfold half life of ¹⁴⁷Nd, the curve obtained was without any maxima not contrasting with the background. Using β -radiation the radioactive purity was examined by constructing the disintegration curves (Fig. 3) 20 days after the interruption of activation when only 147Nd and 147Pm remained unchanged among the initial radionuclides. The disintegration curve obtained by measuring the β -radiation going through the aluminium absorber with a thickness of 0.55 kg m⁻² shows in semilogarithmic coordinates linear character in the whole investigated interval (curve 2). The half life of this disintegration is 11 days, thus it corresponds to pure ¹⁴⁷Nd. On the other hand, the disintegration curve obtained by measuring the non-filtrated β -radiation (curve I) shows a linear drop only in the initial part in conformity with the half life of 11 days (curve 3). Then the rate of activity decrease gradually decreases and when the 1% initial value has been attained, it assumes again a linear character with a relatively high half life of decay. The analysis of this residual radiation by the Feather method using aluminium absorbers with a thickness of 0.026 kg m^{-2} and a standard ¹⁴⁷Pm preparation showed that it was a monocomponent β -radiation with maximum energy of 0.230 ± 0.005 MeV. Such Cp₃Nd preparations, therefore, contained only radioactively pure ¹⁴⁷Pm.

During the irradiation in a reactor Cp₃Nd behaves as a compound fairly resistent to irradiation which may be sublimed in 90-95% yield after irradiation with a dose of γ -rays of about 10³ rad. The retention of ¹⁴⁷Nd in original chemical form expressed as the ratio of specific activity of the Cp₃Nd obtained by sublimation immediately after activation to its specific activity before sublimation was determined by sublimation method (temperature of sublimation 180°C, time of heating about 1 hour). This retention showed the value of 80%.

If the bonds between the daughter atoms of ¹⁴⁷Pm and the cyclopentadienyl rings were impaired in a certain number of cases during the β -decay of ¹⁴⁷Nd bound in the

form of Cp_3Nd molecules and this impairment were irreversible under given conditions, it would result in the stabilization of promethium in a chemical form differing from the form of a tris(cyclopentadienyl) compound. While it may be assumed that the physicochemical properties of Cp_3Pm are similar to properties of Cp_3Nd , other chemical forms of promethium have to be much more different [9, 21]. For instance, the volatility, thermal stability, *etc.* come in the fore in the process of sublimation what will result in a corresponding change in the specific activity of sublimate with respect to the specific activity of sample before sublimation.

Table 1 presents the results of the experiments in which the yield of 147 Pm in the sublimate of Cp₃Nd after the β -decay of 147 Nd was estimated.

The maximum recoil energy of the isolated atom of ¹⁴⁷Pm after β -decay of ¹⁴⁷Nd has the value of 5.3 eV for the maximum energy of β -ray spectrum $(E_{\beta})_{max} = 0.81 \text{ MeV}$ (84%) and 1.8 eV for $(E_{\beta})_{max} = 0.37$ MeV (15%) [19]. In the β -decay of ¹⁴⁷Nd in Cp₃Nd only a certain portion of this energy may be applied to the disruption of the chemical bond between the central atom and the cyclopentadienyl ring. Since the primary product of this process must be the molecular ion $[Cp_3^{147}Pm]^+$ with the electron configuration similar to that of its ground state, the dissociation energy of the isolated ion according to the scheme $[Cp_3Pm]^+ \rightarrow [Cp_2Pm]^+ + Cp$ should be comparable with the dissociation energy of the analogous ion [Cp₃Nd]⁺ the value of which is 1.8 eV (for gaseous phase under the conditions of mass spectrometry at 180°C) [16]. The dissociation involving a splitting off of the second and third cyclopentadienyl rings is not probable from this point of view because it requires the energy of 7-13.8 eV. Though the energetic relations in the solid state are different, in principle, a certain part of primary molecular ions could be transformed in $[Cp_2Pm]^+$ ions by splitting off a Cp radical and the $[Cp_2Pm]^+$ ions might be stabilized in the form of Cp₂Pm. The practical yield of [Cp₂Pm]⁺ ions and Cp₂Pm may, therefore, be to a great extent influenced by the matrix effect (a substantial decrease in recoil energy and a higher probability of recombination) so that the final result of recoil energy may thus become with great probability negligible.

Table 1

Expe- riment	Method of Cp3 ¹⁴⁷ Nd preparation	Period of time between neutron activation and activity measurement [days]	Counting rate before sublimation [s ⁻¹ g ⁻¹]	Counting rate after sublimation [s ⁻¹ g ⁻¹]	Yield of ¹⁴⁷ Pm in sublimate [%]
1a	From activated NdCl ₃	150	143 200	128 200	89.6
16	From activated NdCl ₃	150	145 233	127 766	87.9
2	From activated NdCl ₃	200	123 200	112 000	90.9
3a	Activated Cp ₃ Nd	140	140 850	129 650	92.1
3b	Activated Cp ₃ Nd	140	137 250	125 600	91.6
4	Activated Cp ₃ Nd	160	113 850	106 950	93.9

Yields of $(C_{\epsilon}H_{5})_{3}^{147}Pm$ in the course of the β -decay of ^{147}Nd in tris(cyclopentadienyl)neodymium

TRIS(CYCLOPENTADIENYL)NEODYMIUM

As it follows from the decay scheme of ¹⁴⁷Nd (Fig. 4) a moiety (over 99%) of ¹⁴⁷Pm nuclei at their generation from ¹⁴⁷Nd occur in various levels of excitation [19]. Then they pass into the ground state through converted γ -transitions, the overall coefficient of internal conversion being 0.4. The yield of the fluorescence due to the filling of the vacant places on K level has the value of 88% for an atom with proton number 61 [20]. The remaining 12% is filled under formation of an avalanche of the Auger electrons what results in a high positive charge on central atom. Such multiply charged molecular ions are nearly always subjected to deep fragmentation [21]. From the total number of ¹⁴⁷Nd. disintegrations about 5% proceeds in this way what means that about 5% of the daughter ¹⁴⁷Pm atoms may be stabilized in a chemical form which is quite different from the chemical form of the parent molecules and is not able to sublime under given conditions. Among all possible chemical forms of ¹⁴⁷Pm present in a system only Cp₃Pm will sublime together with Cp₃Nd. Provided a certain part of ¹⁴⁷Pm in the products of stabilization occurred in the form of hitherto hypothetic Cp₂Pm, this substance would remain in the non-sublimed residue. This must be true because it is known that similar compounds of europium and ytterbium sublime in high vacuum in a temperature interval between 400 and 420°C [22]. The distribution of the activity of ¹⁴⁷Pm sublimed together with Cp₃Nd after the β -decay of ¹⁴⁷Nd is on a deaerated chromatographic paper Whatman No. 3analogous to the distribution of the activity of Cp₃Nd labelled ¹⁴⁷Nd [17]. The presence of promethium in the form of Cp₂Pm in a sublimate of Cp₃Nd irradiated with neutrons was also excluded on the basis of the ability to sublime, hydrolyze and the distribution between two liquid phases by Baumgärtner et al. [9]. These data were obtained by investigating the activities of ¹⁴⁷Nd and ¹⁵¹Pm. It is worth noting that in this case the influence of the recoil energy must have been greater because the maximum energy of the β -ray spectrum of ¹⁵¹Nd is 1.93 MeV and therefore the recoil energy of ¹⁵¹Pm is considerably higher than that of ¹⁴⁷Pm.

According to the results given in Table 1, the mean value of the yield of the activity of ¹⁴⁷Pm in a sublimate of Cp₃Nd is 91.3 \pm 3.4%, the probability limit being 0.95. ¹⁴⁷Pm comes into the sublimate in the form of Cp₃Pm which is to a greater extent formed by

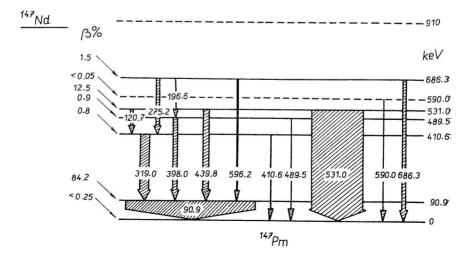


Fig. 4. Decay scheme of ¹⁴⁷Nd.

direct stabilization of that part of primary molecular ions which do not undergo processes of internal conversion or this conversion is not accompanied by the Auger transitions. Such transformations represent about 95% of the total. Since the effect of autoradiolysis as well as the effect of recoil energy on the transition of a certain part of ¹⁴⁷Pm into a non--volatile form was negligible under the conditions used, the processes of internal conversion may be considered to be the major factor in this phenomenon. The processes of internal conversion thus effect that a part of ¹⁴⁷Pm comes to the non-subliming fraction in chemical forms which are not closer defined.

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