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Abstract

Full Text

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Chemistry

Corresponding Member of the Academy of Sciences of the USSR I. N. Plaksin,
L. P. Starchik,
V. T. Tustanovsky

APPLICATION OF THE $(n, 2n)$ REACTION

FOR THE DETERMINATION OF PRASEODYMIUM AND NEODYMIUM

As is known, the principal feature of the rare-earth elements is that they are extremely close in chemical properties. Only a few characteristic chemical reactions are known for certain rare-earth elements. Only cerium, owing to its easy transition to the tetravalent state, can be rapidly and with sufficient accuracy determined colorimetrically.

Recently, the neutron activation method has begun to be used for the analysis of rare-earth elements. Work is known both with the use of a laboratory neutron source of low intensity and with the use of nuclear reactors. Activation analysis of rare-earth elements by the (n, γ) reaction is successfully used for determining trace impurities in ultrapure materials. However, it has the drawback that materials with a large neutron radiative-capture cross section attenuate the neutron flux owing to the self-shielding effect, which leads to obtaining smaller impurity contents than actually exist. To take the self-shielding effect into account, several formulas have been proposed ([1],[2]). However, they are of great value more for establishing the existence of this effect than for determining corrections to the errors resulting from it. Hence the insufficient universality of impurity determination. The determination of praseodymium and neodymium, for example, is ineffective in the analysis of Ce-group raw materials because of the strong influence of samarium (neutron radiative-capture cross section $\sigma_\gamma \mathrm{Sm}^{149} = 40\,800 \pm 3\,000$ barn), europium ($\sigma_\gamma \mathrm{Eu}^{151} = 7\,800 \pm 200$ barn), and gadolinium ($\sigma_\gamma \mathrm{Cd}^{157} = 242\,000 \pm 4\,000$ barn), which are usually present together with praseodymium and neodymium even at late stages of processing.

The disadvantages of this approach to the analysis of rare-earth elements also include its great duration. As a result, rapid control of the processes of enrichment and extraction of the elements is impossible.

Since the capture cross section for 14 MeV neutrons is much smaller than the capture cross section for slow neutrons, it seems of interest to investigate the

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Fig. 1. Spectrum of gamma radiation of Pr140 (0.511 MeV) and Nd141m (0.76 MeV)

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analysis of several rare-earth elements with neutrons of this energy.

In the interaction of 14 MeV neutrons with heavy nuclei, a reaction of the $(n, 2n)$ type is the dominant inelastic process. The behavior of the cross section of the $(n, 2n)$ reaction on heavy nuclei is satisfactorily described by the statistical theory of the nucleus: the magnitude of the reaction cross section in this case is usually greater than one barn. The nucleus arising as a result of such a reaction is an isotope of the target nucleus, which in many cases is radioactive and has a half-life of less than 10 min. The latter circumstance can be used for rapid activation analysis of neodymium, praseodymium, cerium, erbium, terbium, and other elements by the $(n, 2n)$ reaction.

Of particular interest for the control of technological processes is the separate determination of praseodymium and neodymium, which are similar in properties.

Stable praseodymium is a monoisotopic element, and the only possibility for its determination when carrying out activation analysis is the use of Pr^{140} with $T = 3.4$ min, formed by the reaction $Pr^{141}(n, 2n)Pr^{140}$ with a cross section of 2.1 barn. Pr^{140} is a positron emitter with particle energy 2.3 MeV.

For similar cross sections of the $(n, 2n)$ reaction on stable neodymium isotopes: Nd^{142} (27.13%)–2.65 barn; Nd^{148} (5.72%)–2.16 barn; Nd^{150} (5.60%)–

Fig. 1. Spectrum of gamma radiation of Pr^{140} (0.511 MeV) and Nd^{141m} (0.76 MeV)

2.2 barn, only the isomer Nd^{141m} with $T = 64$ sec, $E_\gamma = 0.76$ MeV can be used for rapid activation analysis. In this case lanthanum, europium, and gadolinium, which as a result of the $(n, 2n)$ reaction give longer-lived isotopes, do not influence the determination of praseodymium and neodymium during short-term activation. Cerium present in this group is easily separated from the mixture of elements. The influence of samarium can be taken into account from the energy $E_\gamma = 0.68$ MeV of the isotope Sm^{143m} with $T = 2.3$ min, formed by the reaction Sm^{144} (3.16%) $(n, 2n)$.

In carrying out the analysis, 5-g portions, placed in plastic cassettes 12 mm in diameter, were activated by 14-MeV neutrons using a neutron generator with a flux of $\sim 6 \cdot 10^8$ neutrons/cm$^2 \cdot$sec for 3 min. After irradiation the material was transported to the detector of a scintillation gamma spectrometer and placed in the well of a NaJ(Tl) crystal 40 × 50 mm in diameter. For rapid removal of the sample from the neutron source, a pneumatic-transfer system is usually used.

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Our pneumatic-transfer system delivers the sample from the neutron source to the gamma-spectrometer detector 1 sec after the end of irradiation.

The gamma spectrum obtained after activation of the Pr + Nd mixture is shown in Fig. 1. The spectrum of the induced activity was recorded using an AI-100 analyzer. Praseodymium was identified by the annihilation γ-quanta (E_γ 0.511 MeV) of the β^+-emitter Pr^{140}, and neodymium by the photopeak 0.76 MeV of Nd^{141m}. As can be seen from Fig. 1, the radiation of Pr^{140} is recorded sufficiently clearly using a NaJ(Tl) crystal with a well.

The content of the elements under study was determined by comparing the activity obtained with the activity of irradiated compounds with known contents of praseodymium and neodymium.

The sensitivity of the analysis depends on the flux of fast neutrons, which in our experiments varies from irradiation to irradiation ($10^8 \div 7 \cdot 10^8$ neutrons/cm$^2 \cdot$ sec), depending on the degree of use of the tritium target.

The theoretical sensitivity of praseodymium determination for a flux of $5 \cdot 10^8$ neutrons/cm$^2 \cdot$ sec is $2 \cdot 10^{-6}$ g, and of neodymium $4 \cdot 10^{-6}$ g.

To determine the reproducibility of the analytical results, a series of irradiations of portions with identical contents of the elements under study was carried out, followed by measurement in the region of the corresponding energy peaks. The root-mean-square error of an individual experiment was 4.7%.

Institute of Mining
named after A. A. Skochinskii

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Note: Figure translations are in progress. See original paper for figures.

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