Radiochemical search for the decay of uranium-233 with the emission of neon-24 using a low-background semiconductor gamma-ray spectrometer

A. Ya Balysh, A. A. Gurov, A. V. Demekhin, A. G. Zelenkov, I. V. Kondratenko, B. G. Novatskiĭ, G. A. Pik-Pichak, V. A. Pchelin, Yu. F. Rodionov, L. V. Chistyakov, and V. M. Shubko

I. V. Kurchatov Institute of Atomic Energy, Academy of Sciences of the USSR, Moscow

(Submitted 15 August 1985)
Zh. Eksp. Teor. Fiz. **91**, 37–43 (July 1986)

Radiochemistry and low-background gamma-ray spectrometry were used in a search for the decay of ^{233}U with the emission of ^{24}Ne. The phenomenon was not detected. To a confidence level of 68%, the partial half-life for this effect is greater than 1.7×10^{17} y.

In 1984, a British group discovered[1] a new type of natural radioactivity, namely, the emission of carbon-14 by radium-223. The phenomenon was immediately taken up by several research groups because it occupied an intermediate position between alpha-decay and the spontaneous fission of nuclei. The emission of heavy clusters can therefore provide new information on the structure of the nucleus that cannot be obtained in any other way.

In 1984, the British results on the probability of emission of ^{14}C by ^{223}Ra were confirmed by Soviet[2] and French[3] workers. In 1985, the emission of carbon-14 by the three isotopes of radium with mass numbers 222, 223, and 224 was reported in Ref. 4. Estimates based on the penetration of the Coulomb barrier show that carbon-14 is the most likely to be emitted by the radium isotopes. For protactinium-231 and for the uranium isotopes 232 and 233, the maximum probability is predicted for the emission of neon isotopes.[5] The necessary experiments have been carried out by several groups.[6–9] For uranium-233, it was found that $\lambda(\mathrm{Ne})/\lambda_\alpha \leqslant 7\times10^{-12}$ (Ref. 8) and $\lambda(\mathrm{Ne})/\lambda_\alpha = (7.5\pm2.5)\times10^{-13}$ (Ref. 9). These results were obtained with semiconductor and track detectors of charged particles, which were not capable of determining the mass number of the emitted neon nuclei.

We have therefore repeated the measurements of the probability of emission of heavy clusters by uranium-233, using a different method which enabled us reliably to identify the emerging nuclide. The method involves the radiochemical separation of sodium and a determination of the amount of sodium-24 by high-precision gamma-ray spectroscopy. Sodium-24 was convenient because it has a relatively long half-life (15.02 hours) and emits two cascade gamma-rays (2753.9 and 1368.5 keV) with quantum yields approaching unity.

Thin specimens of the radioactive material must be used to record charged particles. In the radiochemical method, we used about 10 g of uranium-233. Coincidence measurements of the energy spectra of the cascade gamma-rays enabled us to achieve a low background and, consequently, a sensitivity approaching that attained for charged particles.

Neon-24 has a relatively short half-life (3.38 min) and transforms into sodium-24. Strictly speaking, we measured the total probability of emission of neon-24 and sodium-24. We therefore used theoretical estimates of the relative probability of emission of neon and sodium isotopes by uranium isotopes, based on a model similar to that proposed in Ref. 5. However, our calculations were somewhat different. They are presented in the Appendix, and the results are listed in Tables I and II. Table I gives the estimated partial half-lives of uranium isotopes for the emission of neon-24 and neon-25.

The data listed in Table I were obtained for a breakup radius of 1.205 fm. They are very sensitive to this radius. The uncertainty in the absolute estimates is less than two or three orders of magnitude, but the uncertainty in the relative values appears to be less than 0.5 orders of magnitude. Inspection of Table I suggests that the probability of emission of neon-24 is greatest for uranium-232. The probability of emission of neon-24 by uranium-233 is lower by a factor of 100–1000, and the probabilities of emission of neon-24 and neon-25 are very similar.

Table II lists the estimated values of $\log T[c]$ for the emission of neon-24, neon-25, sodium-24, and sodium-25 by uranium-233 for different values of the breakup radius r_0.

It is clear from Table II that the absolute values of the partial half-lives are very sensitive to r_0, but the probability ratios for the emission of different nuclides do not vary much. In view of the uncertainty in the relative measure-

TABLE I.

Nucleus	T, s			
	^{232}U	^{233}U	^{234}U	^{235}U
^{24}Ne	$6.7\cdot10^{23}$	$2.8\cdot10^{26}$	$9.4\cdot10^{28}$	$1.9\cdot10^{31}$
^{25}Ne	$2.1\cdot10^{29}$	$5.3\cdot10^{26}$	$1.4\cdot10^{29}$	$2.5\cdot10^{31}$

 0038-5646/86/070021-04$04.00

TABLE II

Nucleus	r_0, fm		
	1.205	1.25	1.30
^{24}Ne	26.4	24.0	21.3
^{25}Ne	26.7	24.2	21.5
^{24}Na	39.3	36.6	33.7
^{25}Na	33.4	30.8	27.9
^{4}He	14.5	14.0	13.3

ments (0.5 orders), we may conclude that the probabilities of emission of neon-24 and neon-25 are very similar, and the probability of emission of sodium-24 is lower by 12 orders of magnitude and that of sodium-25 by 6 orders of magnitude. It follows that, when we determined the amount of sodium-24 in equilibrium with uranium-233, we were, in fact, measuring the probability of emission of neon-24.

The uranium-233 sample with the lowest concentration of uranium-232 was selected for the experiments. The mass of the specimen was 9.75 g. The sample was dissolved in nitric acid, and the ion-exchange method was used to remove thorium and its radioactive daughter products. The nitric-acid solution of uranium containing 50 mg of the sodium carrier was stored for three to four hours in order to attain radioactive equilibrium with sodium-24. The separation was performed in a hot radiochemical laboratory. This meant that there was an enhanced neutron background due to the presence of radioactive transuranic elements and the proximity of a research reactor. A comparable neutron background could also have been due to (α,n) reactions in the solution (the α-activity of the sample was about 0.1 Ci). Measurements of the neutron activation of sodium with the formation of sodium-24, using a 2.0-g sodium sample placed next to a phial containing the radioactive solution, confirmed our suspicions.

The phial containing the radioactive solution was therefore placed in a borated polyethylene castle with 8-cm walls. Control exposures of 2-g samples of sodium in the castle at the points at which chemical operations were performed, followed by measurements in the low-background gamma-ray spectrometer, showed no evidence of sodium-24 activity.

After the exposure, the nitric-acid solution was passed through the ion-exchange column containing Dowex-1$\times$10 resin (230 mm long and 40 mm in diameter). The column was flushed with a 7 N solution of nitric acid until the sodium was washed out. The eluant was transferred to a clean enclosure, and the liquid was boiled off. The residue was dissolved in hydrochloric acid, the liquid was boiled off, and the residue was roasted. The chemical yield of sodium in these operations was practically 100%.

The sodium chloride was then dissolved in water and a further 500 mg sodium chloride was added to the solution. Intensive radioactive purification of sodium was performed by the repeated removal of barium carbonates, lanthanum hydroxides, lead sulfides, and bismuth sulfides—the daughter products of uranium, thorium, radium and actinium. The chemical yield of sodium in this purification process was 75–82%. The radiochemical operation took an average of 8.5 h to complete.

The sodium chloride samples were sealed in polyethylene ampoules and transferred to the spectrometric laboratory. The gamma-ray spectrometer used to examine the energy spectra consisted of a germanium detector with an efficiency of 20% (measured against a 3$\times$3 inch NaI scintillation counter), surrounded by an NaI phosphor $\sim$8 cm thick. The germanium detector was provided with a channel for inserting samples into it. The semiconductor detector (SCD) had an energy resolution of 2.6 keV at the 1332.5-keV line. The resolution of the scintillation counter at 661 keV was 11%. The detector was provided with a 10-cm shield of low-background lead. The major hazard in our measurements was the nautron background, which could give rise to considerable activation of the NaI phosphor and the sodium specimen in the interior of the SCD. The apparatus was therefore surrounded on all sides with borated polyethylene plates, 10 cm thick. A polymethyl methacrylate phosphor plate was placed on top of the system to protect it from cosmic-ray mesons. The signal from the latter was used to generate a stop pulse which stopped the recording of events for 200 μs ($\sim$4 neutron half-lives in the system) and thus suppressed the fast neutron background accompanying the interaction of muons with the material of the system. The entire system was placed in a cast-iron castle with 140-mm walls (see Fig. 1).

The measurement procedure was as follows. The NaI counter recorded the 2754-keV gamma rays in the interval between 2600 and 2900 keV ($\sim 3\sigma$). These events released a linear gate for pulses from the semiconductor detector. The SCD pulses transmitted by the linear gate (in the absence of the stop pulse from the polymethacrylate plate) then entered a multichannel pulse-height analyzer. The region of interest was the neighborhood of the second gamma-transition of sodium-24 with an energy of 1368.5 keV. Measurements performed on sodium samples irradiated in a research reactor showed that the efficiency of this system of detecting the sodium-24 gamma-rays was 1.4%.

Three experiments were performed. Each series of measurements continued for 15 h. The instrumental background was recorded before and after these measurements (with a sample containing an equal amount of sodium). In the first series of measurements with a 7-keV window, there were no events in the SCD.

In the second series, there was one event (13 h from the start of the measurements) and, in the third series, there

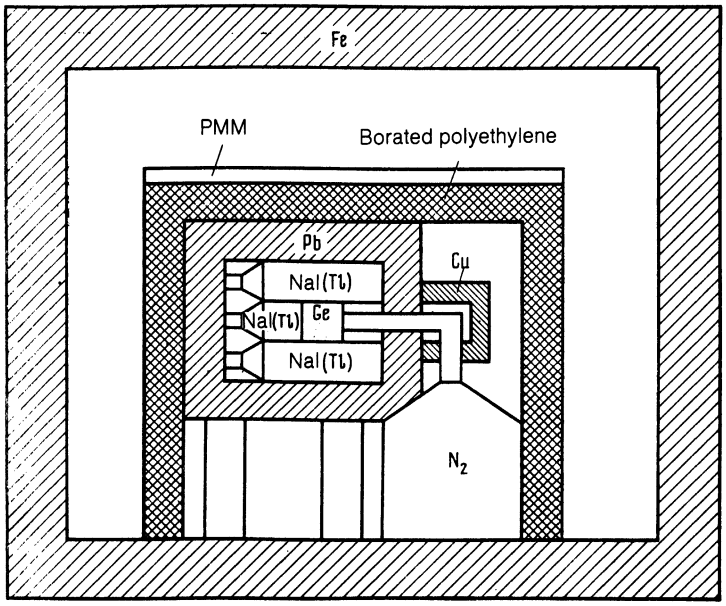

FIG. 1. Apparatus used in the search for sodium-24 gammas.

were two events (after 9 and 14 h from the beginning of measurements). Over 124 h of background measurements, one even was recorded within this window and a further event at the end of the interval (1364.5 keV).

A broad energy interval (between 1268 and 1468 keV) was examined for the gamma-rays recorded by the SCD in order to take correlated coincidences between background events correctly into account. Analysis of the background and the measured gamma-ray energy spectra averaged over all the series (see Fig. 2) showed that the events were distributed quite uniformly. This was used in the mathematical analysis of the measurements. The method of maximum likelihood (MML) was used to look for the desired effect.

The normal distribution of the background-corrected energy spectrum and the corresponding time distribution of the pulse spectrum were taken into account (the experimental data were extracted at hourly intervals).

The required effect has an asymmetric distribution in our case. Within the 68% confidence interval, the effect (e) corrected for the correlated background lies in the range

$$0 \leqslant e \leqslant 0.97 \text{ events/15 h}.$$

Allowing for the detection efficiency (1.4%), the chemical yield ($\sim 80\%$), and the duration of radiochemical operations (8.5 h), we found $N \leqslant 255$ sodium-24 nuclei in radioactive equilibrium with 10 g uranium-233. Hence,

$$T_{1/2}(\text{neon-24}) \geqslant 1.7 \times 10^{17} \text{ y}$$

or

$$\lambda(^{24}\text{Ne})/\lambda_\alpha \leqslant 9.5 \times 10^{-13}.$$

Thus, at the 68% confidence level, our experimental data show that the emission of neon-24 by uranium-233 occurs with a probability that is lower than the values quoted earlier. Our result is not inconsistent with the above theoretical estimates or the data of Ref. 9. The point is that the latter experiment recorded the combined effect of the emission of neon-24 and neon-25 for which the emission probabilities are shown by theoretical estimates to be very similar. On the other hand, we measured only the probability of emission of neon-24. Our method may turn out to be very promising for studies of highly-asymmetric fission by fast charged particles. The nature of this phenomenon is very close to natural radioactivity with the emission of heavy clusters.

APPENDIX

The lifetime of a spherical nucleus for decay into spherical fragments is

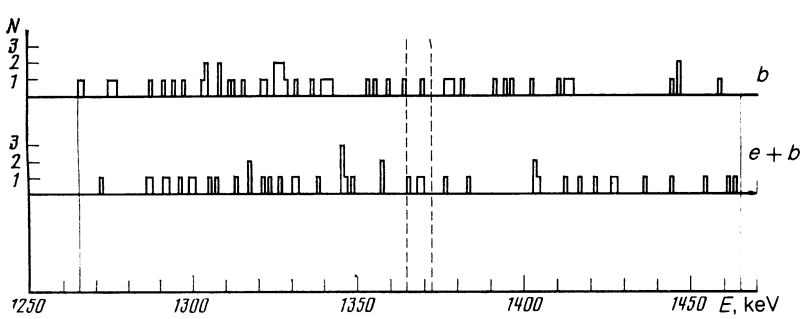

FIG. 2. Background (b) and background plus effect ($e + b$) energy spectra averaged over all measurements (124 h and 45 h, respectively). Broken lines define the region of the expected effect (1365-1372 keV).

$$\log T=\log[2\pi(M_0/C)^{1/2}]+2(S_N+S_C)/\hbar\ln 10,$$

where S_C is the action corresponding to Coulomb forces and S_N the action for Coulomb and nuclear forces,

$$S_C=\int_{R_L^p}^{R_L^c}(2\mu V(R_L))^{1/2}dR_L$$

$$=(2\mu Z_1Z_2e^2)^{1/2}\left\{\frac{\pi}{2\lambda}-\lambda(1-\lambda)^{1/2}-\arcsin\lambda\right\},$$

$$V(R_L)=Z_1Z_2e^2/R_L-Q,\quad \lambda=(QR_L^p/Z_1Z_2e^2)^{1/2},$$

Q is the decay energy equal to the mass difference between the decaying nucleus and the decay products in the ground states, R_L is the separation between the centers of gravity of the fragments, R_L^p is the breakup radius of the fissile nucleus $R_L^p=r_0(A_1^{1/3}+A_2^{1/3})$, Z_1, Z_2, A_1, A_2 are the atomic numbers and mass numbers of the fragments, μ is the reduced mass of the fragments, and $R_L^c=Z_1Z_2e^2/Q$ is the barrier escape radius. Prior to breakup, the shape of the surface of the system is described by the surface of two intersecting spheres of constant density and mass:

$$\rho^2+(z-z_i)^2=R_i^2,\quad A_i=\pi[4R_i^3-3R_id_i^2+d_i^3]/3,\quad i=1,2,$$

where d_i is the height of the spherical segment of radius R_i. The intersection radius is given by $\rho_c^2=d_i(2R_i-d_i)$, and there is no ordered flow of mass through this cross section. The coordinate R_L is given by

$$R_L=\sum_{i=1,2}[R_i-d_i+3d_i^2/4(R_i+d_i)].$$

Prior to breakup, the system experiences Coulomb and nuclear forces, so that the action is given by

$$S_N=\int_{R_L^0+\Delta}^{R_L^p}(2M(R_L)V(R_L))^{1/2}dR_L,$$

where R_L^0 is the initial separation between the centers of gravity of the future fragments in the parent nucleus, which have the shape of a sphere without a segment and a spherical segment, with masses A_1 and A_2, respectively (together they form a sphere of mass $A=A_1+A_2$), $\Delta^2=\hbar/(M_0C)^{1/2}$, and $M_0=M(R_L^0)$. In the Werner-Wheeler approximation,[10] the effective mass for potential flow of a perfect liquid of this configuration is

$$M(R_L)=\pi\rho_m\sum_{i=1,2}\left\{\left(\frac{dz_i}{dR_L}\right)^2\right.$$

$$\times[R_i^2(2R_i-d_i)-(R_i-d_i)^3/3-R_i^3/3]$$

$$+(-1)^i\frac{dz_i}{dR_L}\frac{dR_i^2}{dR_L}(2R_i-d_i)^2+\frac{1}{2}\left(\frac{dR_i^2}{dR_L}\right)^2$$

$$\left.\times[R_i^2/d_i-R_i/2-7(2R_i-d_i)/4-3R_i\ln(d_i/2R_i)]\right\},$$

where ρ_m is the density. For the decay of uranium-233 with the emission of neon-24, $M_0=2.5\mu$ and $M(R_L^p)=\mu$. The potential energy is approximately given by

$$V(R_L)=(E_c-Q+E_v)[(R_L-R_L^0)/(R_L^p-R_L^0)]^2-E_v,$$
$$E_v=\hbar(M_0^{-1}C)^{1/2}/2,\quad C\approx 2(E_c-Q),\quad E_c=Z_1Z_2e^2/R_L^p.$$

It is found that, in fission, the path in S_N is shorter by a factor of 1.4 than in the description of the process in terms of detailed α-decay, as was done in Ref. 5 (the parent nucleus then emits a spherical cluster, i.e., $R_2=$ const during the decay). From Ref. 5, $r_0=1.225$ fm and, to obtain the experimental lifetime for the decay of radium-223 with the emission of carbon-14, the energy of subbarrier motion is increased by $\Delta E=0.128A_2$ MeV, i.e., the conservation of energy is violated. In our case, for radium-223 we have $T_\alpha/T_{^{14}C}=7.3\times10^{-11}$ at $r_0=1.205$ fm and 7.3×10^{-10} at $r_0=1.25$ fm, i.e., the experimental value published in Ref. 1.

[1]H. I. Rose and G. A. Jones, Nature **307**, 245 (1984).
[2]D. V. Aleksandrov, A. F. Belyatskiĭ, Yu. A. Glukhov, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 152 (1984) [JETP Lett. **40**, 909 (1984)].
[3]S. Gales, E. Hourani, M. Hissonnois, *et al.*, Phys. Rev. Lett. **53**, 759 (1984).
[4]P. B. Price, I. D. Stevenson, and S. W. Barwick, Phys. Rev. Lett. **54**, 297 (1985).
[5]D. N. Poenaru, M. Ivascu, A. Sandulescu, and W. Greiner, J. Phys. G; Nucl. Phys. **10**, L183 (1984).
[6]S. Barwick, P. B. Price, and I. D. Stevenson, Phys. Rev. C; Nucl. Phys. **31**, L1984 (1985).
[7]A. Sendulesku, Yu. S. Zamyatin, I. A. Lebedev, *et al.*, Izv. Akad. Nauk SSSR Ser. Fiz. **49**, 2104 (1985).
[8]D. V. Aleksandrov, Yu. A. Glukhov, E. Yu. Nikol'skiĭ, *et al.*, Izv. Akad. Nauk SSSR Ser. Fiz. **49**, 2111 (1985).
[9]S. P. Tret'yakova, A. Sendulesku, Yu. S. Zamyatin, *et al.*, Kratkie soobshcheniya OIYaI (Brief Communications from the Joint Institute for Nuclear Research), 7–85, 23, 1985.
[10]I. R. Nix, Nucl. Phys. A **130**, 241 (1969).

Translated by S. Chomet