ANOMALOUS α-EMITTERS IN THE Po–Ra *REGION*

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Emitters of 7—12 MeV α particles produced by irradiating lead isotopes with accelerated carbon ions have been investigated. Within the limits of experimental error, the half-lives of α-emitters with energies of 7.2 ± 0.2 and 8.8 ± 0.2 MeV are the same (25—26 sec). It is concluded that the radiation is produced in the decay of the Po^{211m} isomer. The half-life of the emitter of 11.7 ± 0.3 MeV α-particles is 47 ± 10 sec. The dependence of the yields of the emitters on the ion energy and nature of the target was determined. On the basis of the experimental data, it is suggested that the 11.7-MeV radiation is due to the decay of a hitherto unknown Em^{214} nucleus in an isomeric state. The α decay of this nucleus is anomalously forbidden. The forbiddenness coefficient is $\sim 10^{14}$.

EMITTERS of 9- and 12-MeV α particles have been observed in experiments described previously.[1] These emitters were produced in reactions in lead by accelerated oxygen and carbon nuclei. According to rough estimates, the half-life is 35 ± 10 sec for the first emitter and ~ 1 min for the second.

It has been suggested that the first group of α particles occur in the decay of Po^{211m}, while the radiation of energy ~ 12 MeV has been ascribed to the decay of an excited At^{213} nucleus produced in the K-capture of a new isomer Em^{213m}. The present experiment was devoted to a more detailed study of the characteristics of the α emitters and their identification.

EXPERIMENTAL METHOD

The experiments were carried out with the internal beam of a 150-cm cyclotron with targets of separated lead isotopes. A monochromatic beam of C^{12} ions of energy ~ 80 MeV was used. To avoid overheating the target, the beam intensity was limited to 0.2—0.3 μA. A special arrangement (probe) similar to that described in [1] was used to record short-lived reaction products. With this arrangement, it was possible to extract the products from the cyclotron chamber within 3—5 sec and bring them to an α detector located ~ 2 m from the target. As the detector, we used NIKFI-T-1 nuclear emulsion and a scintillation α spectrometer with a thin (0.2—0.3 mm) CsI(Tl) crystal. The crystal was introduced into the vacuum section of the probe and the light was brought to an FÉU-11-B photomultiplier through a light pipe 20 cm long. To improve the transmission of the light flashes, the section of the light pipe set against the crystal had a profile in the form of a logarithmic spiral.

The use of a photomultiplier in the direct vicinity of the cyclotron made it necessary to provide magnetic shielding. It is known[2] that the photomultiplier anode current is very sensitive even to weak magnetic fields (~ 0.5 Oe). We used a multilayer shield consisting of two sheaths (steel-3 of thickness 5 and 10 mm) and four insulated layers of tin plate. An investigation showed that when the scintillation counter with such shielding was placed in the leakage field of the cyclotron, the pulse amplitude decreased by 10—15%.

The pulses from the counter were applied to a cathode follower and then through a matched cable 40 m long to a linear amplifier.

In order to eliminate electrical induction at the output of the amplifier, we used a gating circuit which "quenched" the pulses arriving during the application of the high voltage to the dees. The circuit was controlled by high frequency modulation pulses. An integral discriminator which separated the high-energy part of the spectrum for a detailed analysis followed the gating circuit. A 100-channel analyzer AI-100 was used for the amplitude analysis. To illustrate the spectrometric properties of the arrangement, we note that the half-width of the Cm^{242} α lines was approximately 5%.

RESULTS OF THE MEASUREMENTS

1. The α spectra. By improving the technique used previously,[1] we were able to determine

more accurately the energies of the basic group of α particles from the reaction products resulting from interactions of carbon ions in lead. The α particles with energies 7.2 ± 0.2, 8.8 ± 0.2, and 11.7 ± 0.3 MeV were reliably separated from the high-energy part of the spectrum. In some measurements there were indications of the presence of α radiation at ~ 8.3 MeV.

2. Measurement of the half-lives of the high-energy α particles. The determination of the number of pulses recorded in each successive channel of the AI-100 analyzer took, as a whole, only a few minutes. We performed this operation rapidly by photographing the spectrum on the amplitude analyzer screen during the half-life measurements. It was previously established that with a small number of pulses (up to 20) in the channel such "readings" were quite accurate.

The experiment was carried out in the following way. The target was irradiated for 120 sec and then, during ~ 3 sec, the reaction products collector was brought to the crystal counter; during 200 sec, the analyzer was periodically switched on for 30 sec (in a number of runs 40 sec); the spectrum was photographed in the intervals (~ 5 sec) between these periods. After the photography, the analyzer was cleared. In this way, each photograph contained information on the intensity of different groups of α particles during given intervals of time after the end of the irradiation. After the expiration of 200 sec of exposure to the counter, the collector was returned to the target and the cycle was repeated.

Figure 1 shows the results of the measurements summed over 15 cycles. The radiation of ~ 7.2 MeV is shown in Fig. 1a, while the α particles of energy ~ 8.8 and ~ 11.7 MeV are shown in Fig. 1b. The averages over all runs are shown below:

E_α, MeV:	7.2	8.8	11.7
$T_{1/2}$, sec:	25 ± 2	26 ± 4	47 ± 10

It is seen that, within the limits of experimental error, the half-lives of the 7.2- and 8.8-MeV α particles coincide; this confirms the suggestion made previously [1] that the radiation at ~ 9 MeV arises in the decay of Po^{211m}.

3. Dependence of the yield on the energy. In order to obtain the excitation functions, we measured the α-particle spectrum for different energies of the bombarding particles. We varied the energy by moving the target along the radius of the accelerating chamber. The error in the carbon ion energy was estimated as 2—3 MeV. In order

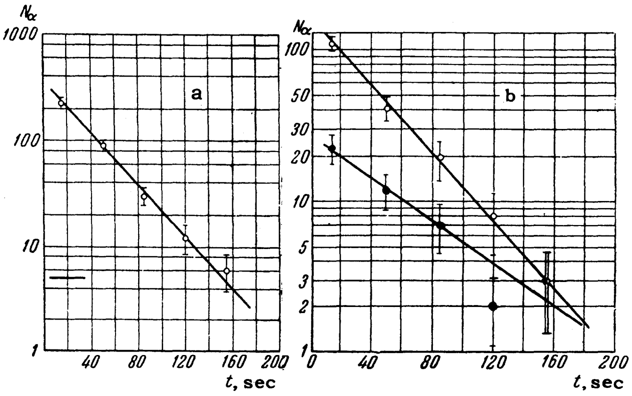

FIG. 1. Variation of α radiation intensity with time: a—7.2 MeV (measured during 30-sec intervals), b—8.8 MeV (○) and 11.7 MeV (●).

to check the correctness of the ion energy measurement (based on the range in aluminum [3]), we determined the energy threshold of the reaction

$$Pb^{207}(C^{12}, 4n)\,Ra^{215} \xrightarrow{\alpha} Em^{211}.$$

It turned out that the experimental value of the threshold (59 ± 2 MeV) differed from the calculated value by less than 1 MeV.

Figure 2 shows the cross sections for the production of emitters of 8.8- and 11.7-MeV α particles when Pb^{208} and Pb^{207} were irradiated* by C^{12} ions. The accuracy of the relative shape of the cross section curve is determined by the statistical errors. The errors for the absolute values depend not only on the statistical errors, but also on the errors in the determination of the ion flux, the effective target thickness, and the effi-

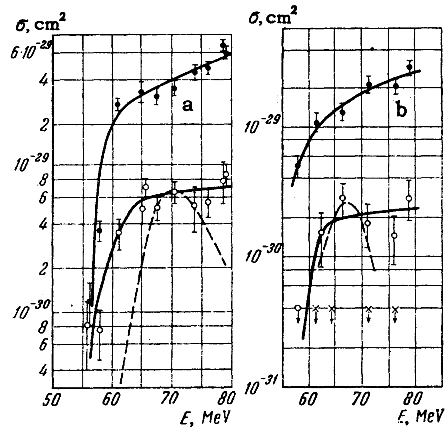

FIG. 2. Variation of cross sections with C^{12} ion energy for reactions leading to the production of α particles with energies 11.7 MeV (○) and 8.8 MeV (●): a—irradiation of Pb^{208}, b—irradiation of Pb^{207}. The crosses with the arrows indicate the upper limit of the cross section for the Hg + C^{12} reaction with the production of an emitter of particles with energy ~11.7 MeV.

*The isotopic composition of the target is given in [1].

ciency of collection and registration of the α-active products. The latter factors lead to an error of ~ 50% in the determination of the absolute value of the cross sections. For the conclusions which we make below, this accuracy is entirely satisfactory; of greater importance is the shape of the curves, which is determined by the type of reaction.

Also shown in Fig. 2b is the upper limit of the cross section for the production of an emitter of α particles with energy ~ 12 MeV upon bombardment of Hg nuclei by C^{12} ions.

For various C^{12} ion energies, we measured the intensity ratio for groups of α particles with energies ~ 7.2 and 8—9 MeV. It turned out that this ratio fluctuates within the limits of 15% about the value 15. It is known that for Po^{211m} this ratio is 13. This result is additional evidence in favor of the conclusion that the α radiation of energies ~ 7.2 and 8.8 MeV is due to the decay of Po^{211m}.

DISCUSSION OF RESULTS

1. In our previous work, [1] it was already suggested that the α radiation at ~ 12 MeV arises in the decay of the nucleus from an excited state (excitation energy ~ 2.5 MeV). Unfortunately, in our experiment we were unable to obtain direct confirmation of this hypothesis; nevertheless, such a suggestion appears to be the only possible explanation for radiation of such high energy.

2. A detailed analysis of the results obtained permits the conclusion that the radiation of ~ 11.7 MeV arises in the decay of an isomeric state of Em^{214}. We shall examine this in detail.

We consider the dependence of the cross sections on the energy of the bombarding particles as shown in Fig. 2. The dotted curves here represent the excitation functions for reactions in which only neutrons (three and two) are emitted. They have been calculated from the formula $\sigma_{xn}(E) = \sigma_c(E)P_{xn}(E)$, where $\sigma_c(E)$ is the cross section for compound nucleus formation, $P_{xn}(E)$ is the probability of the emission of x neutrons at the corresponding excitation energy. [4] The compound nucleus production cross section was taken equal to the fission cross section, which has been measured by Polikanov and Druin. [5] The shape of the experimental curves in Fig. 2 departs considerably from the calculated curves. This indicates that the nuclei under consideration arise in reactions in which not only neutrons, but also charged particles are emitted. A similar form of the excitation function was obtained in [6,7] for reactions in which α particles were emitted. In [7], it was established that the cross section for such reactions does not have a characteristic maximum, it changes weakly in a broad energy interval above the threshold. This, perhaps, is a consequence of the fact that the α particles are emitted in a direct interaction process.

We calculated the thresholds E_t for reactions leading to the occurrence of unknown isotopes of the elements from At to Fr (Ra is excluded from the considerations, since this element arises from the reactions in which only neutrons are emitted). We used here the following relation:

$$E_t = (\Delta E_B + xT_\alpha + yT_p + U^*)(A_1 + A_2)/A_2,$$

where ΔE_B is the change in the binding energy of the system as a result of the reaction; x and y are the numbers of emitted α particles and protons, T_α and T_p are their minimum kinetic energies; U^* is the excitation energy of the residual nucleus in a state from which radiation is emitted at an energy of ~ 11.7 MeV; A_1 and A_2 are the mass numbers of the ion and the target. In the calculations, we used the value $T_\alpha = 15$ MeV based on data for the U^{238} (C^{12}, α4n) reaction[7] and $T_p = 0.5\,B_p = 7$ MeV[8] (B_p is the Coulomb barrier of the compound nucleus for protons). The results of the estimates are given in the table.

For lighter isotopes, the reaction threshold is higher; the heavy isotopes are excluded, since in this case the daughter nucleus should undergo α decay, but in [1] no α radiation genetically connected with the decay of the isotope under consideration was observed. The experimental values of the thresholds for reactions leading to high-energy α radiation are as follows:

Reaction:	$Pb^{208}+C^{13}$	$Pb^{208}+C^{12}$	$Pb^{207}+C^{12}$
Threshold, MeV:	61—62	<60	<62

Comparison of these values with the data of the

Isotope	Reaction	E_t, MeV	Isotope	Reaction	E_t, MeV
Fr^{215m}	Pb^{208} (C^{13}, $p5n$)	81	Em^{214m}	Pb^{208} (C^{13}, $\alpha 3n$)	61.5
Fr^{216m}	Pb^{208} (C^{13}, $p4n$)	75	At^{213m}	Pb^{208} (C^{13}, $\alpha\, p3n$)	74
Em^{213m}	Pb^{208} (C^{13}, $\alpha 4n$)	70	At^{214m}	Pb^{208} (C^{13}, $\alpha\, p2n$)	70
			At^{215m}	Pb^{207} (C^{12}, $He^3\, p$)	73

table indicates that the experimental results are not in contradiction with the suggestion that Em^{214m} is produced in the reactions

$$Pb^{208} + C^{13} \rightarrow Em^{214m} + \alpha + 3n,$$

$$Pb^{208} + C^{12} \rightarrow Em^{214m} + \alpha + 2n,$$

$$Pb^{207} + C^{12} \rightarrow Em^{214m} + \alpha + n.$$

In the calculation of the thresholds shown in the table, the value of U* was based on the energy of the most favorable scheme for nuclear decay. For Em^{214m}, we considered the following scheme

$$Em^{214m} \xrightarrow[T_{1/2} \approx 47 \text{sec}]{\alpha\,(11.7\text{ MeV})} Po^{210}.$$

Other possible schemes for the Em^{214m} decay (K-capture and β^- decay in an excited nucleus with subsequent emission of a high-energy α particle) correspond to thresholds higher (68–70 MeV) than that measured in this experiment. Moreover, a detailed analysis of these schemes shows that the energy of the Em^{214m} isomeric state should then be ~ 9–10 MeV, i.e., greater than the binding energy of a neutron.

Figure 3 shows the most probable Em^{214m} decay scheme. The probability of a radiative transition to the ground state was estimated from the

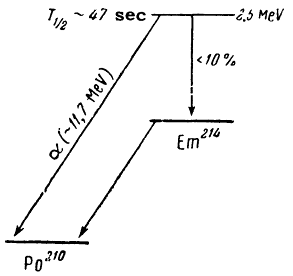

FIG. 3. Most probable decay scheme leading to α radiation of energy ~11.7 MeV.

relative intensities of the α particles with energies ~ 8.8 and ~ 11.7 MeV, so that part of the first group can be attributed to the Em^{214m} decay from the ground state. The minimum value of this ratio was 0.1 (determined from the data of [1]). It then follows that the partial lifetime of Em^{214m} for the γ transition, is not less than 500 sec.

3. The Em^{214m} α decay is anomalously forbidden. Calculations according to semiempirical formulas in good agreement with experiment (see [9]) indicate that the half-life for an even-even Em nucleus in case of an energy ~ 11.7 MeV should be ~ 2×10^{-13} sec. It follows from this that the forbiddenness coefficient for Em^{214m} α decay is anomalously large, about 10^{14}. This value is 10^5 times the largest known forbiddenness coefficient (for Po^{211m}).

We note that the conclusion on the high forbiddenness for high-energy radiation essentially does not depend on the identification of the radiation. We shall consider, for example, a decay scheme suggested in our earlier work according to which the α radiation is associated with the decay of Em^{213m}:

$$Em^{213m} \xrightarrow[T_{1/2} \approx 47 \text{sec}]{K} At^{213*} \xrightarrow[T_{1/2} - \text{small}]{\alpha(11.7\text{MeV})} Bi^{209}.$$

Em^{213} is an α-active nucleus with a decay energy from the ground state of ~ 7.8 MeV (according to the estimates in [10]). If K-capture leading to an At^{213} nucleus excited to ~ 2.5 MeV is to occur with $T_{1/2} \approx 1$ min, it is necessary to assume that the Em^{213m} excitation energy is at least 6 MeV. In this case, α decay with energy ~ 13.8 MeV competes with the Em^{213m} K-capture. The absence of this radiation indicates a forbiddenness not less than 10^{17}.

The anomalously high forbiddenness coefficient for Em^{214m} α decay can be due to the isomeric state of this nucleus having a complex structure, i.e., connected with excitation of several nucleons. The probability for the production of α particles from the nucleons in essentially different states is considerably reduced. Moreover, an additional slowing down of the speed of decay can be connected with the large angular momentum of the Em^{214m}; from an estimate of the partial lifetime for the γ transition, it follows that the Em^{214m} spin is $I \geq 5$.

4. The α radiation of energy ~ 7.2 and ~ 8.8 MeV arises in the Po^{211m} decay. In favor of such a conclusion is the agreement between the energies, half-lives, and relative intensities of the lines.[11]

In the interaction of carbon ions with lead, Po^{211m} is produced as a result of the following reactions

$$Pb^{208} + C^{13} \rightarrow Po^{211m} + 2\alpha + 2n,$$

$$Pb^{208} + C^{12} \rightarrow Po^{211m} + 2\alpha + n,$$

$$Pb^{207} + C^{12} \rightarrow Po^{211m} + 2\alpha.$$

In conclusion, we note that the present work does not at all satisfy our interest in the emitter of the 11.7-MeV α particles. Further investigations of the properties of this nucleus are necessary.

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Note added March 13, 1962. The 11.7-MeV radiation arises, according to our suggestion, in the decay of Em^{214m}. This most probable identification, however, is not unique. The table gives estimates of the reaction thresholds leading to only unknown isotopes. It turns out that the calculated reaction thresholds leading to new isomeric states of the known isotopes Po^{211} and Po^{212} do not differ greatly from the experimental ones. Therefore, strictly speaking, we cannot exclude the possibility that the 11.7-MeV radiation arises in the decay of new Po isomers. We note once again that the basic conclusion of this work is the detection of a new α activity of an isomer with an anomalously high forbiddenness coefficient for α decay. This conclusion does not essentially depend on the details of the identification of the high-energy particle emitter.

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