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On the effect of chemical bonds on the β-decay of Tritium and ^{241}Pu

(Translation of two reports)

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BETA-DECAY OF TRITIUM INTO BOUND CHEMICAL STATES AND ITS EFFECT ON THE ACCURACY OF CALORIMETRIC MEASUREMENTS*

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ABSTRACT

The effect of ß-decay into bound chemical states of the daughter atom on the half-life of ß-active nuclei with a low maximum transition energy is evaluated. The probability of ß-decay into a bound chemical state strongly depends on the electron environment of the daughter nucleus, and for tritium this can represent up to 1% of the total decay probability. The accuracy of the calorimetric method in measuring ß-active nuclei with a low maximum energy depends on how accurately the probability of ß-decay into the continuous spectrum is known. It is recommended that the degree of uncertainty for the tritium half-life be increased and be taken as 12.34 ± 0.06 years instead of 12.35 ± 0.01 years.

[Key words: tritium, half-life, effect of chemical bond.]

The development of a technique capable of determining low thermal powers with an accuracy of about 0.1% would seem to enable amounts of radioactive substances to be determined with good accuracy [1]. If a mean energy $\bar{E}$ is released during the decay of one nucleus and if all that energy is absorbed inside the calorimeter, then, once the thermal power W of the radioactive sample has been determined and the decay constant is known, it is not difficult to obtain the number of radioactive nuclei N in the sample from the equation

$$N = \frac{W}{\lambda \bar{E}} \qquad (1)$$

This method can be applied to those radioactive substances whose radiation is absorbed inside the calorimeter.

The aim of this paper is to draw attention to the need, when using Eq. (1) for $ß^-$-activity with a low $ß^-$-ray spectrum maximum energy (for example, tritium and plutonium-241), to take into account the chemical and bound state of radioactive atoms which influences the decay probability. In Eq. (1), λ must be understood as the probability of ß-decay with electron emission into the continuous spectrum and not as the total ß-disintegration probability.

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* Translation of the Kurchatov Atomic Energy Institute preprint IAE-3102

The problem is that for ß-decays with a low maximum energy there is a high probability of electron capture in the shell of the atom being formed, though this process cannot be recorded by the calorimeter since practically all the decay energy is carried away by the antineutrino.

The expression for the thermal power of the sample which takes account of this effect is written as:

$$W = N(\lambda_\beta \bar{E}_\beta + \lambda_e \bar{E}_\gamma), \qquad (2)$$

where λ_β is the probability of decay with electron emission into the continuous spectrum; λ_e is the probability of decay in which the electron is captured by the atom; $\bar{E}_\beta$ is the mean energy of ß-particles; $\bar{E}_\gamma$ is the mean energy of the optical transitions which accompany the electron capture process. The second term in Eq. (2) is much smaller than the first term, as normally $\lambda_e \ll \lambda_\beta$ and $\bar{E}_\gamma \ll \bar{E}_\beta$, and so the product $\lambda_e \bar{E}_\gamma$ in Eq. (2) can be disregarded.

Let us examine the case of tritium. According to the international Evaluated Nuclear Structure Data File (ENSDF), its half-life is 12.35 ± 0.01 years. This value is in good agreement with the independent evaluation carried out by the International Nuclear Data Centre of the State Committee on the Utilization of Atomic Energy (GKAE) [2]. The mean energy of the emitted electrons, according to ENSDF data, is 5.68 keV and can easily be calculated from the known transition energy using the ß-decay theory equations.

However, these data do not take account of the effect of the chemical and bound state of tritium. In order to evaluate the extent of the possible influence of the above-mentioned effects on tritium half-life, two models will be studied: a fully-ionized tritium atom model and a neutral atom model.

Similar models have frequently been studied by various authors [3]. However, the results of their work are in poor agreement with each other which may be due either to misprints in publications or to inaccuracies on the part of the authors.

Let us examine the ratio between the probability of tritium ß-decay into the discrete spectrum P_B, in which the electron is captured into the 1S state of the helium atom and the probability of tritium ß-decay into the continuous spectrum P_c [4]:

$$\Delta = \frac{P_B}{P_c} = 2\pi\alpha^3 \left(\frac{Q}{mc^2}\right)^2 \frac{Z_{eff}}{f} \sim \frac{105}{8\sqrt{2}} \alpha^3 \left(\frac{mc^2}{Q}\right)^{\frac{3}{2}} Z_{eff}^3, \qquad (3)$$

where $$f = \int_1^{1+Q/mc^2} F \cdot \left(1 + \frac{Q}{mc^2} - E\right)^2 E \sqrt{E^2-1}\, dE \sim \frac{16\sqrt{2}}{105} \left(\frac{Q}{mc^2}\right)^{7/2},$$

$$F = \frac{1}{2p^2}\left(g_{-1}^2 + f_1^2\right)$$

g_{-1} and f_1 are the radial components of the emitted electron wave function; Q is the maximum energy of the ß-ray spectrum; P is the electron momentum; α is 1/137.03604; E is the ß-particle energy.

The effective helium charge Z_{eff} in our calculations can be taken as 27/16 [5] for a filled tritium atom shell, and as 2 for an unfilled shell. The values of the function F were taken from tables calculated by Suslov in Ref. [4]. A numerical calculation gives $\Delta_{ion} = 9.1 \times 10^{-3}$ for ionized tritium decay, and $\Delta_{at} = 5.5 \times 10^{-3}$ for a neutral tritium atom. If the probability of electron capture into excited states of helium is taken into account, then $\Delta_{ion} = 11 \times 10^{-3}$, and $\Delta_{at} = 6.6 \times 10^{-3}$.

Thus, the probability of tritium ß-decay with electron capture into the helium shell can attain approximately 1% of the total ß-decay probability and be negligibly small when there is a large electron concentration near the decaying nucleus. The chemical environment, of course, also affects the probability of tritium ß-decay, with electron emission into the continuous spectrum. According to Alder et al. and to Wilkinson [6], $\lambda_ß$ is 0.3% greater for ionized tritium decay than for neutral tritium atom decay. Thus, λ_e clearly makes a considerable contribution to the correction to the total half-life due to the interreaction of the electron with the medium. It follows from the statements above that the effect of the chemical environment and bound state is of such magnitude that it is essential to take it into account for accurate measurements (of the order of 0.1%). A theoretical calculation of the decay probability for actual substances requires a detailed knowledge of the local field intensity of the atom, hence, given the present status of theory and experiment, it is easier to measure Δ than to calculate it. Unfortunately, no experiments have yet been carried out to measure $\lambda_ß$ or λ_e.

The following procedure is one possible way to carry out this measurement. A sample with an accurately known quantity of tritium must be prepared; the

thermal power released by it and its time dependence must then be measured. Since in Eq. (2) $\lambda_e \overline{E}_\gamma << \lambda_\beta \overline{E}_\beta$, then as we know the number of tritium atoms and the thermal power, λ_β can be calculated and, from the time dependence, $\lambda_\beta + \lambda_e$ can be determined. The proposed experiment is virtually a combination of the method described in Ref. [7], in which a quantity of tritium and helium-3 was accurately determined, and the calorimetric methods described in Refs [1, 8].

In our view, a larger uncertainty value for the tritium half-life than is recommended in ENSDF must be assumed, i.e.: for tritium, $T_{\frac{1}{2}}$ should be taken as 12.34 ± 0.06 years. Similar effects for plutonium-241 and possibly even for carbon-14 should also be taken into account in practice.

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COMMENT ON THE HALF-LIFE OF ^{241}Pu*

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ABSTRACT

The authors evaluate the extent to which beta decay accompanied by capture of the electron formed into the $7s_{\frac{1}{2}}$ state of the daughter atom can influence the total lifetime of ^{241}Pu. The probability of decay via this channel is strongly dependent on chemical bonds. It is shown that the upper limit of the fraction of decay accompanied by ß-electron capture may be 50%.

In spite of the high accuracy achieved in various experiments for measuring the lifetime of ^{241}Pu the available experimental data do not agree sufficiently closely with each other [1]. However, a knowledge of the nuclear physics characteristics of ^{241}Pu is important because it is a fissile product that accumulates during the operation of nuclear power plants. The existing scatter in the experimental data, although not large in absolute terms, is important with respect to applications, as the proceedings of various experts' meetings show (see, for example, Refs [2, 3]).

This scatter in the half-life data, we believe, can be explained by the influence of chemical bonds. As we showed in an earlier paper [4], the probability of ß-decay with capture of the electron formed into the atomic shell of the daughter nuclide is strongly dependent on chemical bonds and, under certain conditions, may constitute a large fraction of the total probability of ß-decay.

For tritium this fraction is 1%. The lower the energy output in ß-decay and the stronger the overlap of the wave functions of the nucleus and of the electron captured by the daughter nuclide atom, the greater the influence of this decay channel on the observed half-life. In the case of ^{241}Pu, assessing the influence of chemical bonds is complicated in particular by the fact that this decay mode is a first forbidden ß-transition.

Nevertheless a determination of the extent of the influence of capture of the electron formed by the nuclide ^{241}Am may be attempted.

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* Translation of an informal note from the authors.

The ratio of the probability of decay with electron capture by the atomic shell, P_β, to the probability of decay with escape of the ß-electron from the atom, P_c, takes the form:

$$\frac{P_\beta}{P_c} = \frac{2\pi^2 C_{N_i} \varrho_e(Am)\,(E_o - \Delta)^2}{f}$$

where ϱ_e is the square of the modulus of the wave function of the captured electron, Δ is its binding energy, E_o is the energy output in decay, $f = \int_1^{E_o} C_N F(E,Z)\sqrt{E^2-1}E(E_o - E)^2 dE$ is an integral Fermi function, C_N is the ß-spectrum profile factor, and C_{N_i} is a quantity analogous to C_N but relating to ß-decay in a discrete spectrum. In the case $E_o \ll m_e c^2$, $f \sim C_N \frac{\sqrt{216}}{105} E_o^{\frac{1}{2}}$ and $P_\beta/P_c \sim E^{-3/2}$.

Since the dependence of C_N and C_{N_i} on the energy and the nuclear matrix elements is complex in the case of forbidden ß-transitions, P_β/P_c does not take as simple a form as it does in the case of tritium. But the maximum value to be expected can be obtained.

The energy output in ^{241}Pu decay is 20.8 keV $\ll m_e c^2$, and the valence electrons are in the $7s_{\frac{1}{2}}$ state. In the range E < 20 keV C_N is practically independent of E [5]. Then, taking C_N from under the integration sign and then multiplying and dividing by $\varrho_e(^3He)$ and $f(^3He)$, we obtain the relation

$$\frac{P_\beta}{P_c} = \frac{2\pi^3 (E_o - \Delta)^2 \varrho_e(^3He)}{f_o(^3He)} \cdot \frac{C_{N_i}}{C_N} \cdot \frac{\varrho_e(Am)}{\varrho_e(He)} \cdot \frac{f_o(^3He)}{f_o(Am)}$$

The first multiplier in this expression is P_β/P_c for tritium, equal to ~1%.

$$\frac{\varrho_e(Am) f_o(He)}{\varrho_e(He) f_o(Am)} \sim 100 \qquad [6]$$

If we assume that $C_{N_i}/C_N \sim 1$ as in the case of forbidden transitions, then we obtain for ^{241}Pu $P_\beta/P_c \sim 100\%$. Of course C_{N_i}/C_N is less than unity and its value can also be determined by accurate machine calculations. But from all the foregoing we may conclude that the decay in the discrete spectrum of ^{241}Pu is appreciable, and may be significant for ^{241}Pu ions. This in turn leads to the dependence of the ^{241}Pu lifetime on chemical bonds, which may possibly also be responsible for the scatter in the experimental data on the half-life of ^{241}Pu.

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