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No Decelerated β -Decay of Tritium Solved in Titanium

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Experimental results on the interaction of tritium with fine particles of titanium, published in Phys. Lett. A and attributed to decelerated β -decay of tritium solved in titanium, are instead explained by adsorption of tritium on the surface of the particles, formation of tritium oxide when oxygen is present, and normal β -decay.

In one of the latest issues of Physics Letters, O. Reifenschweiler, retired Chief Physicist of Philips Research Laboratories, published a paper [1] on "Reduced radioactivity of tritium in small titanium particles", describing research work done at Eindhoven about 30 years ago. In the discussion of the experimental results, two states of tritium are mentioned: The gaseous state and tritium absorbed in the lattice of the titanium particles. In no case, however, the existence of a third state of tritium, i.e. as adsorbate at a solid surface, for instance at Ti, has been taken into account. That this state played an enormes role in the experiments becomes clear from a simple calculation.

The process of producing the titanium particles has been described [1] in the following way: "In our experiments the titanium preparation was made by evaporation of the metal in argon at a suitable pressure, e.g. 0.5 to 2 cm Hg. It was deposited as a kind of soot, consisting of monocrystalline particles about 15 nm in diameter, arranged in chains, on the inner wall of the measuring vessel. ... After the argon was pumped out, the tritium was added and was completely absorbed within a few seconds."

In the experiment described first, a layer of 48 mg Ti was prepared in this way. The applied quantity of Ti, 10.8 mm^3 (density $\varrho_{\text{Ti}} = 4.43 \text{ g cm}^{-3}$) equals 6.5×10^{15} spherical particles of 15 nm diameter, presenting a surface of 4.15 m^2 . A monolayer on this surface corresponds to about 4.15×10^{19} tritium atoms, or 0.07 mmol. An amount of 48 mg Ti, on the other hand, is nearly 1 mmol Ti. Therefore, a monolayer of tritium

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atoms on the surface of the small Ti particles corresponds to a compound of $TiT_{0.07}$. Since this experiment was done with $TiT_{0.0035}$, the capacity of a monolayer on the Ti surface was by about a factor 20 higher (!).

The course of this investigation was, to add $0.0035 \times 1/2$ mmol T₂ to the layer of 1 mmol Ti single crystals at room temperature, and then to heat up the system – under continuously pumping out – to temperatures just above 400 °C. The β -activity of the tritium was determined by measuring the X-radiation, accompanying the β -decay (bremsstrahlung and characteristic X-rays) by a GM-tube. The number of GM-counts per second is

$$n = \alpha \lambda N_{\rm T} \tag{1}$$

where N_T is the number of tritium atoms, λ their decay constant, and α the counting efficiency of the GM-tube, which was at the beginning of the experiment determined to be $\alpha = 4.2 \times 10^{-9}$.

In Fig. 1 the counts per min are plotted versus the temperature T of the system, showing in curve A the results of this heating experiment. Curve B displays the pressure of the released tritium gas when heating up the system under closed condition. There occurs a reduction of the counting rate from the initial value down to 72% at $160\,^{\circ}$ C and to 60% at $275\,^{\circ}$ C. Thereafter the counting rate comes up again to the initial value at about $360\,^{\circ}$ C, and then falls down steeply to the background value. O. Reifenschweiler interprets this reduction of the counting rate as a decrease of the decay constant λ in (1) and develops for this a nuclear pair hypothesis [1] which he himself, indeed, characterizes as "highly unorthodox".

Taking into account the adsorbed state of tritium, the explanation of Fig. 1 can be given as follows. A few seconds after adding tritium into the gas room of the system at normal temperature, the tritium is adsorbed at the titanium surface, the count rate and the efficiency, α , have their initial values. These values stay constant, until around 115 °C tritium atoms begin to enter the lattice of the Ti particles. The distribution of the tritium now changes by diffusion, and the counting efficiency decreases owing to the difference of the bremsstrahlung in the Ti lattice compared to the free gas room above the Ti surface, and owing to the absorption of the X-radiation by the Ti. The reduction of the counting rate, attributed by O. Reifenschweiler to

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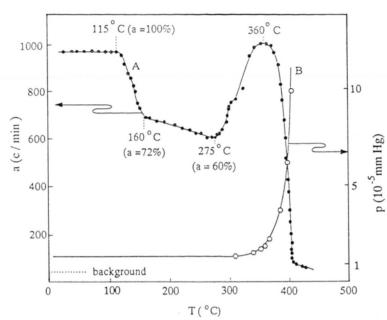


Fig. 1. Graph A: Count rate as a function of temperature in the pumped system. Graph B: Tritium pressure as a function of temperature in the closed system (reproduced from [1]).

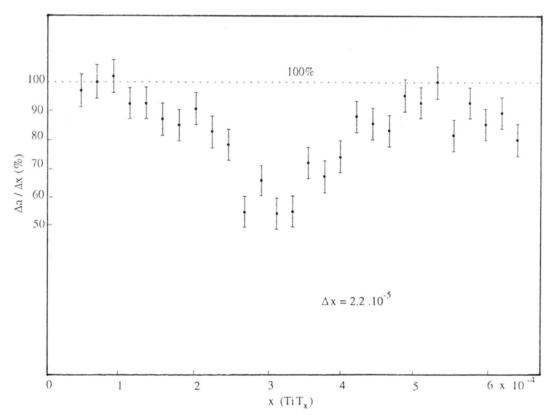


Fig. 2. Increase of radioactivity $\Delta a/\Delta x$ as a function of concentration x. Evaporator removed (reproduced from [1]).

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a decrease of the decay constant λ of the tritium, is explained here by a decrease of the counting efficiency α as a result of the change in the distribution of the tritium.

At temperatures higher than about 300 $^{\circ}$ C the Ti-T-system begins to decompose, T₂ molecules are released from the Ti surface, and the radioactivity, after having recovered shortly the initial value, falls off steeply as the consequence of the loss of tritium.

The results of a second series of experiments, made isothermally at room temperature by adding successive quantities Δx of tritium to TiT_x systems [1], can also be explained by the activity of adsorbed layers of tritium. In this case, only 3.6 mg of Ti were deposited as finely divided particles on a gold layer at the inner wall of the measuring glass vessel, and the β -decay of the tritium was followed by a special electrometer device.

The amounts of tritium, added successively in these TiT_x experiments, were very small ($\Delta x = 2.2 \times 10^{-5}$), see Figure 2. Of prime importance in this case is, however, that the evaporator of the Ti was removed from the glass vessel prior to the first addition of tritium, and on this occasion impurities, especially oxygen,

will have had access to the freshly deposited Ti layer. The first additions of tritium, adsorbed at the surface of the small Ti particles, will form OT-groups at the centers of this oxygen impurity, but will display full radioactivity in the electrometer, see the left side of Fig. 2 (100%). The next additions Δx of tritium will form more and more T₂O; this will desorb from the Ti surface and migrate to the glass wall, where T₂O is more strongly adsorbed than at a metal surface. By this migration, however, parts of the tritium will no more be catched fully by the electrometer device. Therefrom the decrease in the measured growth of radioactivity of the later additions of tritium, down to about 55% at $x = 3 \times 10^{-4}$, see Figure 2. After further additions of tritium, the Ti surface will be more and more free of oxygen, hence the successive increase of the measured radioactivity, Fig. 2, up to the initial value of 100%.

In this way, all somewhat puzzling results of the different series of experiments described in [1] can be explained by well-known and normal effects, and it seems not to be necessary to develop a new hypothesis on any change in the decay constant of tritium.