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Some Experiments on the Decrease of the Radioactivity of Tritium Sorbed by Titanium

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Abstract

A sharp decrease of the radioactivity of tritium was observed when the hydrogen isotope is sorbed by small monocrystalline particles of titanium and the preparation is heated to several hundred degrees centigrade. In other experiments the concentration of tritium in such preparations was varied, showing that the radioactivity of the tritium increased less than proportionally to its concentration. A first attempt is presented to explain these remarkable effects in terms of a "nuclear pair hypothesis".

1. Introduction

In a recent letter [1] the author has given a short description and preliminary interpretation of experiments where a sharp decrease of the radioactivity of tritium was observed. The present paper describes these and additional experiments in more detail. By this procedure a high degree of evidence was obtained for the strange effect of the decrease of the radioactivity of tritium. I should like to recall that the experiments have been done many years ago in the course of technological projects.

2. Experimental

As described in ref. [1] titanium preparations were made by evaporation of the metal in argon at a pressure of 1 up to 2 cm Hg. By homogeneous nucleation small monocrystalline particles of titanium are obtained which are arranged in chains with many ramifications. By this procedure a very loose soot-like deposit is obtained. After pumping out the argon tritium is introduced and is sorbed with a time constant of about 10 s.

Two different measuring procedures are applied to determine the radioactivity of tritium. In one part of the experiments, the heating experiments described in this paper, a thin stainless steel or nickel window enables measurement of the x-radiation accompanying the β-decay by a GM tube (fig.1). In some other experiments the β-emission current is measured directly by a vibrating reed electrometer. While the first measuring procedure is applied for thick preparations, e.g. on an average about 80 Ti-particles thick, the second one can only be applied for thin preparations, about one particle thick. An analysis of the measuring procedures is given in ref. [1].

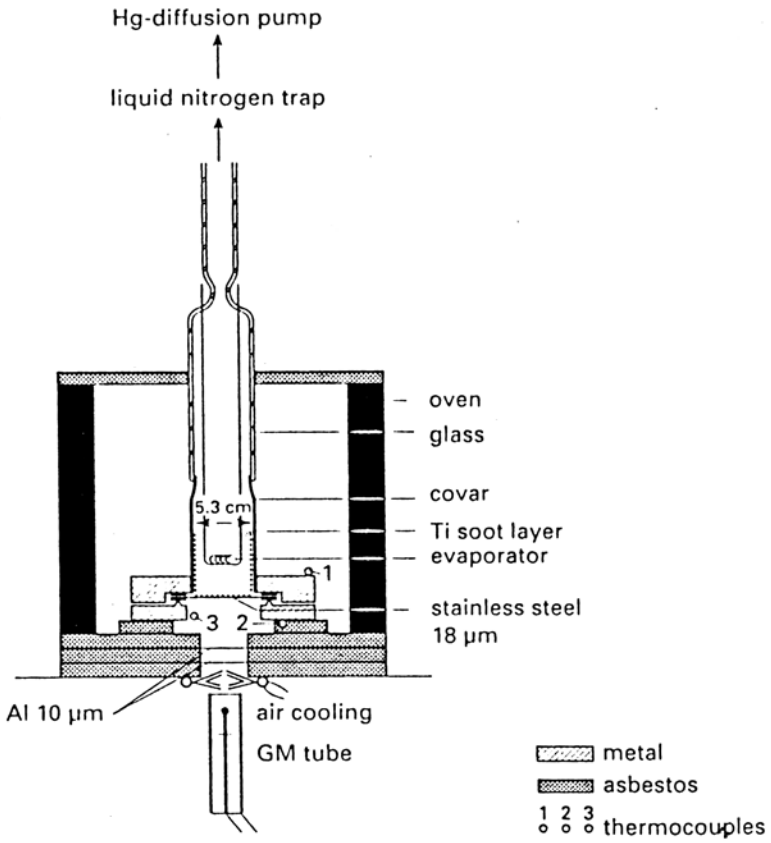

Fig. 1. Experimental arrangement for measuring the β-radioactivity as a function of temperature via the x-radiation by a GM-tube.

3. Heating Experiments

The heating experiment presented in ref. [1] is described and evaluated in more detail in this paper.

In a preliminary experiment the pressure rise of tritium by heating a $TiT_{0.0035}$-preparation (48 mg Ti, 100 mC T_2) twice in a closed system was determined. Fig. 2 shows the result. With a first rise of temperature the tritium pressure (graph B) shows no increase at temperatures below about 350°C and then increases. With the second rise of temperature the tritium pressure (graph C) increases very much earlier at about 250°C. A possible explanation of this important phenomenon may be that the release of the tritium at the first rise of temperature is prevented by surface contamination [2], above all titanium oxide, which is dissolved in the metal during the heating up to 480°C at the first rise of temperature. This preliminary explanation has to be carefully investigated applying UHV-technique and definitely determined oxide layers. Such investigations should support our preliminary conclusion that the pressure rise upon the second rise of temperature indicates the release of tritium atoms from their bonds to the titanium lattice.

In a most important heating experiment [1] an identical preparation is heated in the

arrangement shown in fig. 1. Fig 2 graph A gives the count rate as a function of temperature. As stated in ref. [1] it is very strange that the count rate decreases sharply between 115°C and 160°C by 28% followed by a further slower decrease, reaching 60% of the initial value at 275°C and then rises to about the initial value at 360°C. With a further increase of temperature the count rate decreases fast, which is due to decomposition of the preparation.

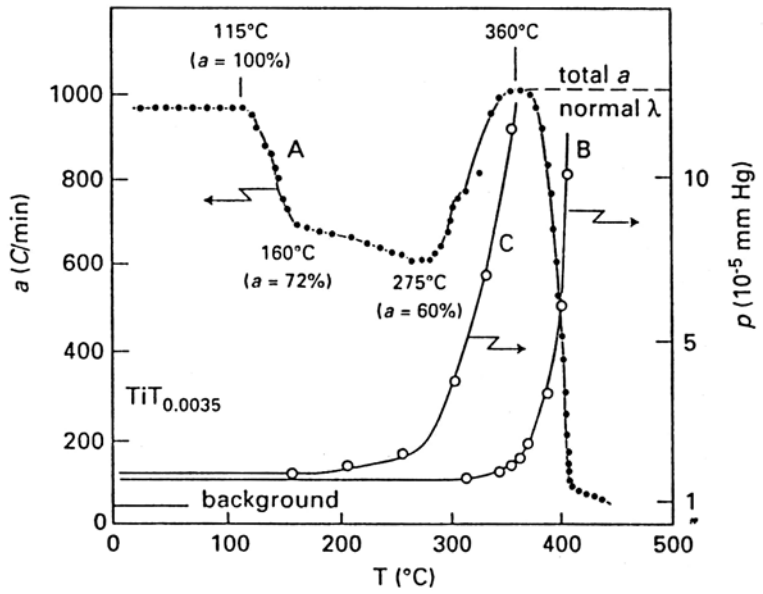

Fig. 2. As a function of temperature, TiT0.0035-Preparation:
Graph A: Count rate, pumped system.
Graph B: Tritium pressure at the first rise of temperature, closed system.
Graph C: Tritium pressure at the second rise of temperature, closed system.

The sharp decrease of the radioactivity above all between 115°C and 160°C, but also the further slower decrease between 160°C and 275°C, cannot be explained by decomposition of the preparation. This is clearly shown by graph B which represents the increase of the tritium pressure and hence the release of tritium upon a first rise of temperature: in such a preparation there is no measurable release of tritium below about 350°C, which is confirmed by many experiments. But the strongest argument that no tritium is lost at the first decrease of count rate between 115°C and 275°C is the fact that the radioactivity increases again between 275°C and 360°C to the initial value before decomposition of the preparation. Thus we arrive at the following preliminary conclusion:

In the temperature region between 115°C and 275°C a new compound of tritium is formed with a lower λ_T and with further increase of temperature this lower emitting compound is destroyed.

The re-increase of the count rate begins at the same temperature as the increase of tritium pressure at the second rise of temperature. This leads to the preliminary conclusion that the binding of the tritium atoms (nuclei) to the titanium lattice is a necessary condition for the

decrease of the radioactivity. However the tritium atoms released from their bonds to the titanium lattice which exhibit normal λ cannot leave the titanium particles, as shown by graph B.

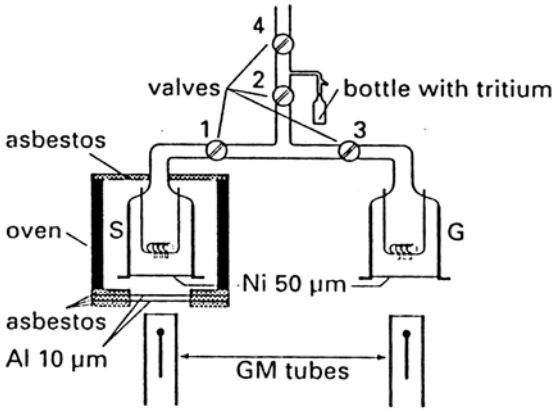

Fig. 3. Experimental arrangement with two tubes. Tube S contains a $TiT_{0.035}$-preparation at the beginning of the experiment and is heated in an oven. Tube G contains a finely divided Ti-preparation which absorbs the tritium released from preparation S.

In a further heating experiment a preparation with a 10 times higher concentration of the tritium (48 mg Ti, 1 Ci tritium) is investigated. In this experiment the tritium is desorbed from the $TiT_{0.035}$-preparation during heating on account of the higher concentration. Fig. 3 shows the experimental arrangement. The tritium which is released from the preparation in tube S (solid) is absorbed by an identical titanium preparation in a second tube G (gas) maintained at room temperature and the radioactivity is measured by a second GM tube. Fig. 4a shows the result. Graph S gives the count rate of tube S which is heated and graph g that of the second tube G remaining at room temperature which determines the amount of the released tritium gas. The count rate of tube S is constant up to about 210°C and then decreases steeply. This led to the preliminary conclusion that the tritium is desorbed from preparation S during heating.

To proceed to a more complete evaluation of this experiment we normalize the count rate of tube G to the same sensitivity as count rate S (fig. 4b). The count rate G gives the radioactivity of the tritium gas released from tube S and S+G is a measure of the total radioactivity. As can be seen a similar decrease and re-increase of the total radioactivity takes place as in fig. 2, and the minimum of the radioactivity is attained at the same temperature of 275°C. If we attribute the decrease of the radioactivity only to the solid preparation of tube S we obtain graph S'.

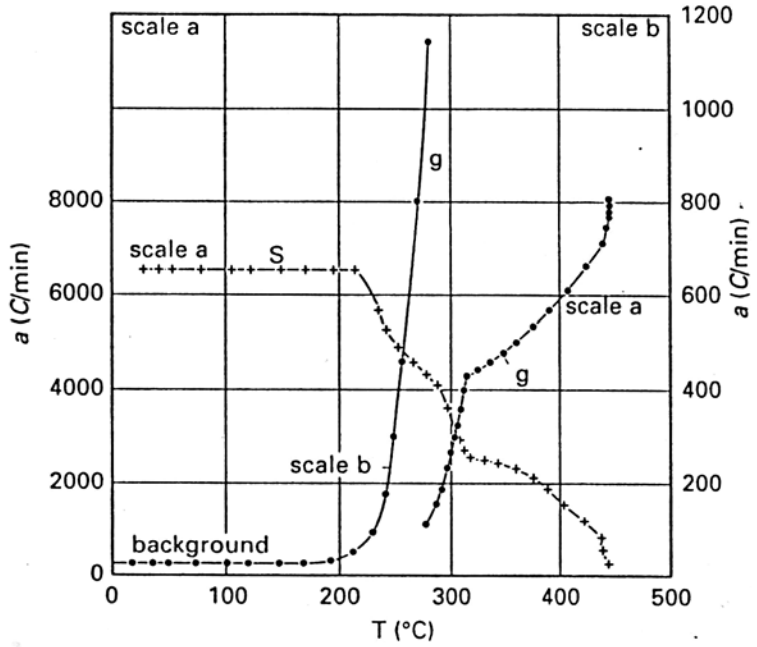

Fig. 4a. Count rate as a function of temperature
S: for tube S
g: for tube G

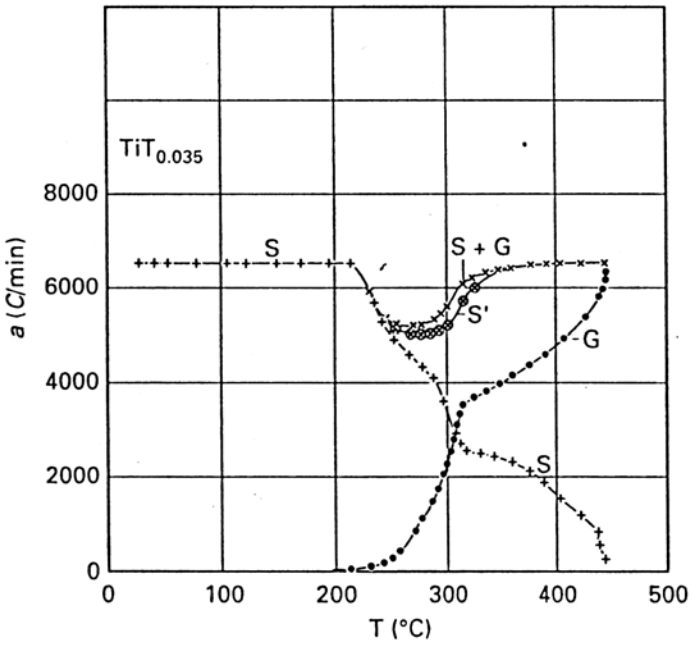

Fig. 4b. Radioactivity as a function of temperature
S: for tube S
G: for tube G
S+G: total radioactivity
S': remaining radioactivity in the preparation of tube S

Most interesting is the evaluation of the decrease of the radioactivity above 210°C. Between 210°C and 230°C the decrease of the radioactivity is 12.5 times higher than the amount of the released tritium and between 210°C and 242°C the corresponding value is 10.5 times higher. Thus the strong decrease of the radioactivity S above 210°C cannot be caused by the release of tritium from the preparation. This is a further strong argument for the decrease of the radioactivity during heating of our preparation above 210°C. That is just the beginning of the pure α-phase [3]. Then monocrystallinity of the small particles is regained which was disturbed by extremely small hydrid particles precipitated in the small TiT-α phase particles.

It can be shown that the course of a = f(T) for the two heating experiments done under quite different conditions is equivalent. By evaluation of our first heating experiment we arrived at the preliminary conclusion that T-atoms (nuclei) must be bound to the titanium lattice to get a decrease of the radioactivity. In the first experiment (fig.2) the tritium atoms released from their bonds to the titanium lattice cannot leave the small titanium particles probably on account of surface contamination, above all oxidation. They have normal λ, and the re-increase of the radioactivity above 275°C is due to these liberated tritium atoms remaining in the preparation. With the second experiment (fig 4) the tritium atoms released from their bonds to the titanium lattice leave the small titanium particles on account of the higher concentration. The total radioactivity re-increases in the same manner as with the first experiment. The only difference is that with the first experiment the tritium atoms which regain normal λ remain in the preparation and with the second experiment they leave the small titanium particles as a gas on account of the higher concentration.

It is most important to apply an oxide layer to the titanium particles or another means to prevent release of tritium from the Ti-particles in a heating experiment of the kind described in ref. [1] and fig.2 of this paper when working with UHV technique. It may be possible to distinguish between bulk absorption and chemisorption at the surface of the Ti-particles by performing experiments with a different degree of surface oxidation. It is recommended to apply in all experiments a second tube or another means to determine a possible release of tritium gas from the preparation.

Two other heating experiments, where the surface of the TiT_c-particles is oxidized by exposing the preparations to ambient air before heating, also distinctly show the effect. However the diminution of the radioactivity was not so strong as with the “clean” preparations (fig. 5). Especially interesting is the experiment with a $TiT_{0.065}$-preparation, with a tritium concentration nearly twice as high as that of the experiment where the tritium was desorbed during heating. In this experiment the tritium remained in the preparation until the re-increase of the radioactivity occurred. Obviously the release of tritium was prevented by a strong oxide layer on the surface of the titanium particles. It seems furthermore that the oxidized surface layer (about 4 nm) of the small Ti-particles does not participate in the decrease of λ_T .

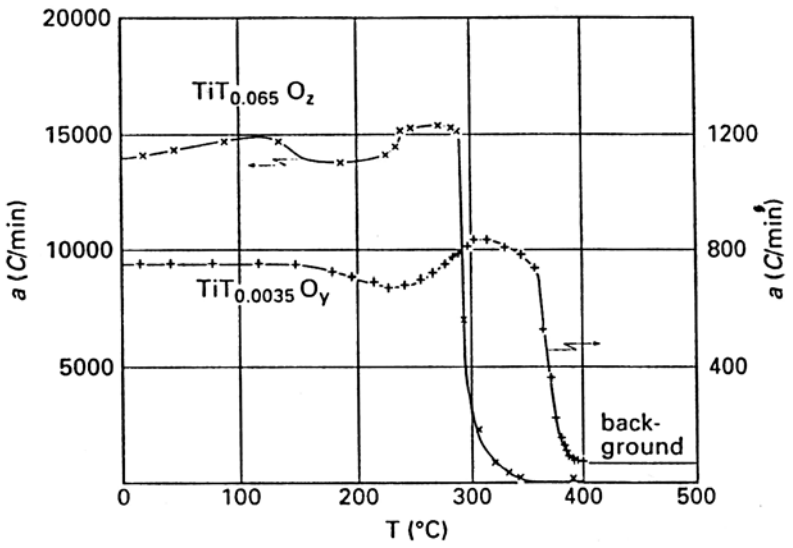

Fig.5. Count rate as a function of temperature for oxidized preparations.

The release of the tritium atoms from their bonds to the titanium lattice can be prevented by maintaining an equilibrium pressure of tritium above the preparation. With this mode of operation we may expect a further decrease of the radioactivity. Such experiments provide confirmation or our preliminary conclusion that the binding of the tritium atoms to the titanium lattice is a necessary condition for the decrease of the radioactivity.

Up till now in all heating experiments the formation of the lower emitting compound occurred in the α-phase of the titanium-tritium system, thus with very low concentrations. It is extremely interesting to perform such experiments with higher tritium concentration, with the hydride phase TiT_c.

For a theoretical interpretation of the effect it may be important that the tritium atoms (nuclei) are bound in harmonic oscillator potential wells.

In a recent opposing note E. Wicke [4] argued that the decrease of the radioactivity *a* in fig. 2 is caused by migration of the tritium atoms from chemisorption at the surface to bulk absorption when the radioactivity decreased and conversely that it returns to the surface upon the re-increase of the radioactivity. However the effect proposed in ref. [4] cannot explain the course of $a = f(T)$ presented in figs. 2 of this paper and of ref. [1]. On average about 80 Ti-particles are piled up in our titanium layer and the β-electrons are either completely stopped in the layer, or those which leave the layer enter it at another part of the tube wall. Thus the Ti layer and the upper part of the tube wall supporting the layer are a homogeneous source of x-radiation whose intensity is proportional to the β-radioactivity of the preparation. Furthermore Wicke's argument cannot apply to the second heating experiment where the concentration of tritium strongly decreased during re-increase of the total radioactivity. A contra-note will be submitted for publication in the near future.

4. TiT_x-Experiments

By our heating experiments we arrive at the conclusion that by heating TiT_c-preparations (c=0.0035; 0.035 and 0.065) a new compound of the tritium in the small titanium particles with a lower λ_T is formed. However there is no physical effect known that can account for the observed decrease of radioactivity.

It has been speculated that the decrease of λ_T in the small titanium particles may have something to do with the different phases of the TiT_c-system (a suspicion which has so far not been confirmed). Therefore experiments were undertaken to measure the radioactivity as a function of the concentration in such systems. The concentration was taken as independent variable x and the relating experiments are therefore indicated as TiT_x-experiments. In these experiments small accurately determined quantities of tritium were added successively to a thin finely divided titanium preparation and the increase of the emission current Δi was measured by a vibrating reed electrometer. The result of the most important experiment is shown in fig. 3 of ref. [1]. With increasing x $\Delta a/\Delta x$ first decreased reaching a minimum at $x \approx 3\times10^{-4}$ and then increased again to the initial value at $x \approx 5\times10^{-4}$.From this and further TiT_x-experiments a "nuclear pair hypothesis" was derived which reads explicitly:

Two tritium nuclei (identical nuclei with half integer spin) under certain conditions (in our case by embedding in small particles of titanium) arrange themselves in pairs with nuclear spin zero. Such a nuclear pair acts for the decay to a certain extent as the parent nucleus, so that the radioactive decay changes to a certain extent into a higher forbidden one.

The author would like to emphasize that the "nuclear pair hypothesis" is a *hypothesis*. This implies that it is a first attempt to explain the strange effect of the decrease of λ_T. It is to be proved or disproved by further experiments. Furthermore nothing has yet been said about the nature of the hypothetical nuclear pairs.

5. Concluding Remarks

There is a strong suspicion that the two different effects, cold DD-fusion and decrease of tritium radioactivity are caused by the same or a related fundamental principle: hydrogen isotopes, D or T, are sorbed in suitable metals and nuclear properties are changed in a quite incomprehensible manner. It should be investigated if cold DD-fusion happens in preparations as used in our experiments at elevated temperatures. If so thermodynamic analysis may lead to a deeper understanding of the effect. The fact that sometimes cold fusion experiments with a positive result are not reproducible points to the existence of a hidden necessary condition. For the effect of λ-decrease of tritium such a condition may be monocrystallinity or another may be smallness of the metal particles. Both can be investigated experimentally. Also compact layers have to be investigated.

It may be interesting that in the experiment of M. Fleischmann and S. Pons [5] where excess enthalpy production was observed with deuterium absorbed in palladium analogous conditions for the formation of small $PdD_{0.6}$-particles (hydride phase) are present as for the formation of our monocrystalline particles: Increasing the concentration of deuterium above the end of the α-phase ($PdD_{0.01}$) one gets supersaturation of the α-phase, which results inhomogeneous nucleation. This brings about formation of small monocrystalline particles of Pd-hydride. To get homogeneous nucleation the Pd-metal has to be very pure, a condition which is

also necessary for the production of excess enthalpy as reported by the authors of ref. [5]. Otherwise one gets inhomogeneous nucleation with the formation of bigger and non-monocrystalline hydride particles. It is suggested that the microstructure and possible monocrystallinity of the palladium hydride suspended in the Pd-matrix should be investigated by metallographic techniques at different states of saturation.

With both effects, cold DD-fusion and decrease of tritium radioactivity, identical particles interact. The cold DD-fusion gives the possibility to check if such interactions are only possible between identical particles (indicated as "homo-interactions") or also between non-identical ones (indicated as "hetero-interactions"). If one gets neutron production in a cold fusion experiment by DD-reaction one has to repeat the experiment with a mixture of e.g. 50% deuterium and 50% tritium. If the reaction occurs between non-identical particles one has then to observe a high intensivity of 14 MeV neutrons from the DT-reaction. The result of such an experiment is very important for the theoretical interpretation of the cold fusion effect. If the reaction happens only between identical particles many theories are ruled out.

Acknowledgments

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