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Reduced radioactivity of tritium in small titanium particles ☆

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By heating a $TiT_{0.0035}$ preparation consisting of extremely small monocrystalline particles ($\phi \approx 15$ nm) a decrease of the radioactivity by 40% was observed. In further experiments the concentration of tritium in such preparations was varied (TiT_x experiments) showing that the radioactivity of the tritium increased less than proportionally to its concentration. Careful analysis of the experiments seems to rule out the possibility of trivial errors. A provisional hypothetical explanation is formulated. Our experiments may point to a connection with cold DD-fusion.

Several recent publications deal with "cold fusion" of deuterons suitably absorbed in solid matter (e.g. Pd or Ti) [1–3] and although some difference of opinion may still exist as to the interpretation of the results, they have certainly stimulated renewed interest in the question whether nuclear phenomena can be influenced by the atomic and electronic environment. In this connection the author wants to report some curious results obtained many years ago in the course of technological projects [4–8] based on the use of the hydrogen isotope tritium absorbed in titanium [9–11]. It seems to me that it would certainly be worth repeating these experiments with up-to-date experimental technology, but unfortunately I am no longer in a position to do this myself [#1]. Hence this communication.

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 [4,6]. 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. In fig. 2 of ref. [4] and fig. 1 of ref. [6] electron micrographs of such preparations are shown. After the argon was pumped out the tritium was added and was completely absorbed within a few seconds. This was confirmed by hundreds of experiments. In one part of the experiments the electron current (β-particles and secondaries) was measured by a vibrating reed electrometer via a cylindrical electrode placed inside the vessel. In several other experiments there was a thin steel window to enable the x-radiation accompanying the β-decay (internal and external bremsstrahlung and characteristic X-rays [12]) to be measured by a GM-tube (fig. 1). Various experiments established for both detection systems the linear relationship between the read-out and the activity.

In one experiment [5] a $TiT_{0.0035}$ preparation (48 mg Ti, 100 mC T_2) is slowly heated (duration about 10 h) in the arrangement of fig. 1 and the radioactivity is measured by the x-radiation. The tritium which is released – if any – is pumped out continuously. Figure 2, graph A gives the count rate as a function of temperature. The increase of tritium pressure during the heating of an identical preparation is determined in a separate experiment in a sealed-off tube (graph B). It is very strange that the radioactivity a decreases sharply between 115°C and 160°C by 28% followed by a slower decrease, reach-

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[#1] A more detailed description of our experiments with proposals to improve the experimental technique can be obtained on request from the author.

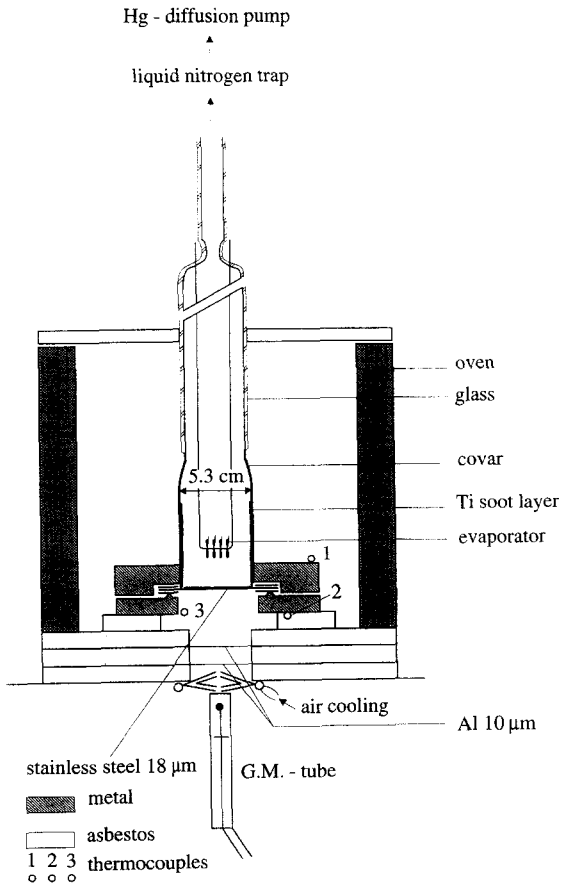

Fig. 1. Experimental arrangement for the relative measurement of the β-activity as a function of temperature measuring the x-radiation by a GM-tube.

ing 60% of its initial value at 275°C, and then rises steeply to the initial value at 360°C (graph A). With further increase of temperature the count rate decreases fast due to decomposition of the preparation and hence the loss of tritium. The first decrease of radioactivity between 115°C and 275°C, however, cannot be explained by decomposition of the preparation. Graph B shows that there is no measurable release of tritium below about 300°C (confirmed by many experiments). But the strongest evidence that no tritium is lost at the first decrease of radioactivity between 115°C and 275°C is the re-increase of the count rate between 275°C and 360°C.

In two other experiments carried out under different conditions a similar decrease and increase of

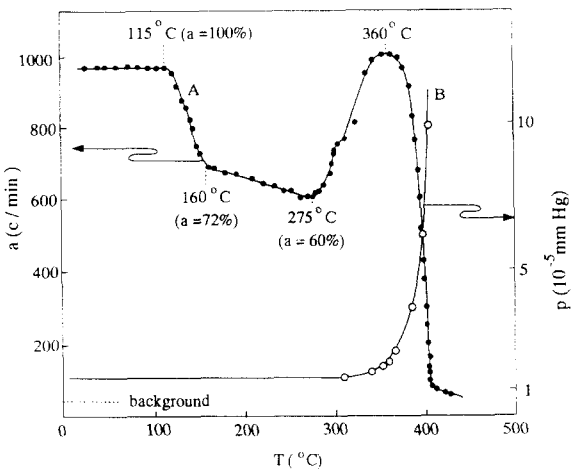

Fig. 2. Graph A: Count rate as a function of temperature in the pumped system. Graph B: Tritium pressure as a function of temperature in the closed system.

the radioactivity with increasing temperature was observed. But there was also one experiment with a 10 times higher concentration of tritium and a roughly 5 times faster increase of temperature where the decrease of count rate did not occur.

To arrive at an explanation of the strange result presented in fig. 2 we proceed in the following way: If n is the number of GM-counts per second, N_T the number of tritium atoms, λ the decay constant and α the counting efficiency, then we have

$$n = \alpha \lambda N_T .$$

As was explained above, we are convinced that the decrease of n between 115°C and 275°C cannot be caused by a decrease of N_T. The counting efficiency α comprises the efficiency of generation of internal and external bremsstrahlung and of characteristic X-rays by the β-electrons of tritium [12], the overall geometrical factor (only a very small part of the generated X-rays hit the GM-tube) and the counting efficiency of the GM-tube. The generation of internal bremsstrahlung is independent of external conditions and thus proportional to the number of β-electrons. A change of the efficiency of generation of external bremsstrahlung, of characteristic X-rays and of the overall geometrical factor between 115°C and 275°C would require a considerable migration of tritium, although – as was explained above – no tritium

was escaping from the titanium layer. Such a migration seems thus impossible. The counting efficiency of the GM-tube was tested several times and may be assumed constant during the experiment. Thus it is hard to see that the counting efficiency α could change at temperatures between 115°C and 275°C. It is calculated that $\alpha = 4.2 \times 10^{-9}$ at the beginning of the heating experiment.

So we are almost forced to assume a decrease of λ between 115°C and 275°C during our experiment even though no physical effect is known that brings about such a high change of radioactivity [13].

In order to obtain further evidence for a change of λ another series of experiments was made at room temperature. The concentration x of the tritium was varied in a finely divided TiT_x preparation and the corresponding increase of the emission current caused by the β-electrons of tritium was measured (TiT_x experiment). In these experiments a substantial deviation from the expected proportionality between current i and concentration x was observed: $\Delta i/\Delta x$ is not constant with increasing concentration x but shows characteristic variations. In one of these experiments 3.6 mg Ti was deposited as a finely divided preparation with particle size of about 16 nm on the inner wall of the measuring vessel (a bulb of 10 cm diameter covered inside by a Au layer). After removing the evaporator a cylindrical electrode of 5 cm diameter and a length of 8 cm was lowered into the centre of the bulb, the system was sealed off and a negative voltage $U = -252$ V was applied to the Au electrode carrying the TiT_x preparation. With this voltage the $i = f(U)$ curve (current–voltage characteristic) was nearly horizontal. The small accurately determined quantities of tritium were added successively and the increase of the current $\Delta i/\Delta x$ was measured. By recording the tritium pressure using a special tube which measures the β-emission of the gaseous tritium it was ascertained that the tritium has been absorbed completely after each addition. Figure 3 shows the result. With increasing x $\Delta i/\Delta x$ first decreases reaching a minimum of about 55% of its initial value at $x \approx 3 \times 10^{-4}$ and then increases again to the initial value at $x \approx 5 \times 10^{-4}$.

Again we can write the number of the detected β-electrons per second n determined from the emission current as

$$n = \delta \lambda N_T \quad \text{or} \quad i = \delta i_\beta ,$$

where i is the measured current and i_β the β-emission current.

The number of tritium atoms N_T absorbed in the preparation and with it Δx and x were carefully determined at each addition of tritium as stated above. The counting efficiency δ contains a possible absorption effect of the β-electrons in the titanium and the overall geometrical factor consisting of the loss of electrons not hitting the central electrode and the addition of backscattered and secondary electrons from the Au backing of the TiT_x preparation. The mean thickness of the titanium preparation was determined as 25 nm, which is about the range of 1 keV electrons [14]. Thus the absorption effect of the titanium is very small. Furthermore from several observations a uniform distribution of the tritium in the Ti preparation is concluded, the small absorption effect does not change during the experiment and is thus negligible. Furthermore we cannot see any reason why the overall geometrical factor should depend on x. We thus come to a constant counting efficiency δ during the experiment. At the beginning of the experiment it was determined as $\delta = 0.89$. We may thus conclude that the measured current i is proportional to the β-emission current of the tritium in the preparation and the increase of the measured current $\Delta i/\Delta x$ may be replaced by the increase of the radioactivity $\Delta a/\Delta x$ in the preparation.

There was also an experiment in which the radioactivity was determined by the x-radiation as in our heating experiment and where a course of $\Delta a/\Delta x = f(x)$ of the same kind as that shown in fig. 3 was obtained.

So we are once more forced to the conclusion that we have to look for a decrease of λ of the added tritium. However, as stated above with the heating experiment such a marked change of λ is never observed experimentally [13] and unfortunately there is no theory that can describe such a pronounced decrease of λ. It may be that current theory has overlooked some effects that might occur because of the rather special conditions: emitted electrons have to find their place among the electrons of extremely small single crystals. But it seems unlikely that this can cause a 40% decrease of λ in out heating experiment and also the roughly 45% decrease of λ of the

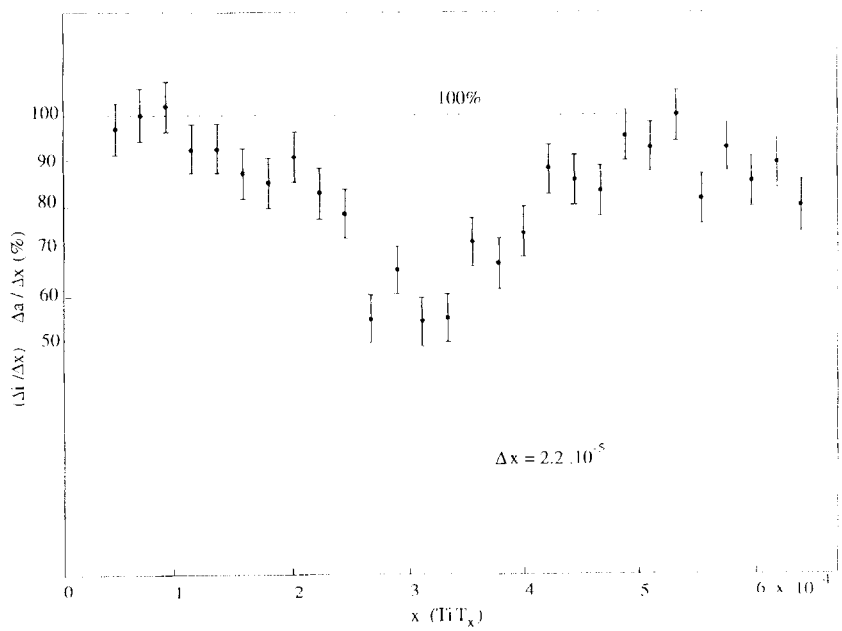

Fig. 3. Increase of radioactivity $\Delta a/\Delta x$ as a function of concentration x. Evaporator removed.

added tritium at the minimum of the $\Delta a/\Delta x = f(x)$ curve.

Even more puzzling are the following results: In two independent experiments $\Delta a/\Delta x$ was found to decrease at $x \approx 3 \times 10^{-3}$ to very low values of a few percent of the initial value. This observation, however, is not quite certain because the evaporator was not removed from the measuring vessel and could have absorbed some tritium. But during the course of these experiments several additions of tritium were followed by a decrease of the radioactivity ($\Delta a/\Delta x < 0$), which even continued when all tritium was absorbed. This extremely strange behaviour cannot be explained by absorption of tritium by the evaporator.

Under these circumstances it seems justified to put forward a highly unorthodox hypothesis, the nuclear pair hypothesis. If we assume that tritons absorbed in the extremely small single Ti-crystals can combine into pairs and that the decay constant of such a pair is much smaller than that of a free triton, then the observed behaviour of all TiT_x experiments can be explained.

The author – though well aware that the experimental evidence is rather limited and that a theoretical foundation is lacking – feels strongly attracted by this idea of nuclear pairing with reduced radioactivity and he believes that it might have other applications. The author hopes to come back to these questions in later publications.

It should be obvious that our results might also have a bearing on cold fusion [1–3]. As a first step I should like to propose experiments with deuterium absorbed in preparations of finely divided hydrogen absorbers (Ti, Pd or others) as used in our tritium experiments and at temperatures between 100°C and the dissociation temperature.

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References

[1] M. Srinivasan, Curr. Sci. 60 (1991) 417.
[2] E. Storms, Fusion Technol. 20 (1991) 433.
[3] M. Fleischmann and S. Pons, Phys. Lett. A 176 (1993) 118.

[4] O. Reifenschweiler, in: Proc. Conf. on Peaceful uses of atomic energy, vol. 19 (United Nations, Geneva, 1958) p. 360.
[5] J.F. van de Vate, O. Reifenschweiler and G. van de Ligt, Onderzoekingen naar de temperatuurvastheid van fijnverdeelde metaal-tritium-systemen in vacuüm, Internal report, Philips Research Laboratories (1961), unpublished.
[6] O. Reifenschweiler, Rev. Sci. Instrum. 35 (1964) 456.
[7] H. Hörster, H. Lydtin, O. Reifenschweiler and K.G. Fröhner, Z. Angew. Phys. 22 (1967) 203.
[8] O. Reifenschweiler, in: Proc. 1968 Int. Conf. on Modern trends in activation analyses, Special Publication 312, Vol. II (National Bureau of Standards, Washington, DC, 1969) p. 905.
[9] W.M. Mueller, in: Metal hydrides, eds. W.M. Mueller, J.P. Blackledge and G.G. Libowitz (Academic Press, New York, 1968) p. 336.
[10] J.R. Morton and D.S. Stark, Trans. Faraday Soc. 56 (1960) 351.
[11] G. Alefeld and J. Völkl, eds., Hydrogen in metals, Vols. I and II, Topics in applied physics (Springer, Berlin, 1978).
[12] D.K. Wickenden and O. Reifenschweiler, Some investigations into the bremsstrahlung of tritium, Internal report, Philips Research Laboratories (1961), unpublished.
[13] G.T. Emery, Annu. Rev. Nucl. Sci. 22 91972) 165.
[14] J.E. Holliday and E.J. Sternglass, J. Appl. Phys. 30 (1959) 1428.