ABOUT THE POSSIBILITY OF DECREASED RADIOACTIVITY OF HEAVY NUCLEI

NUCLEAR REACTIONS IN SOLIDS

KEYWORDS: *radioactivity, gas-solid exchange reactions, diffusion experiments*

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Received September 11, 1995
Accepted for Publication July 24, 1996

Recently a sharp decrease in the radioactivity of tritium was reported, and a preliminary explanation of this effect was formulated in terms of a nuclear-pair hypothesis. Through the evaluation of several gas-solid exchange and diffusion experiments of others, where heavy radionuclides (^{65}Zn, ^{63}Ni, ^{85}Sr) are used as tracers, it can be shown that such an effect may also exist for these nuclei. In all these experiments the second law of thermodynamics seems to be grossly violated. By pure formal application of the nuclear-pair hypothesis, all such deviations from normal behavior can be explained. Several straightforward experiments are proposed to prove the decrease in radioactivity of heavy nuclei.

I. INTRODUCTION AND OUTLINE OF THE PROBLEM

In a recent letter[1] a sharp decrease in the radioactivity of tritium was claimed when the hydrogen isotope was sorbed in small monocrystalline particles of titanium, after this preparation had been heated to several hundred degrees centigrade. Evaluation of further heating experiments[2,3] performed under quite different conditions provides a large amount of evidence supporting the existence of this strange effect. In further experiments the concentration of tritium in such preparations was varied at room temperature (indicated as TiT_x experiments), showing that the radioactivity of the tritium increased less than proportionally to its concentration. A first attempt to explain these remarkable effects in terms of a nuclear-pair hypothesis[1-3] was presented. I should like to remind the reader that the relating experiments were performed ~30 yr ago in the course of technological projects.

The question arises as to whether such an effect may also exist for nuclei heavier than tritons (indicated as heavy nuclei). Not being in a position to perform relevant experiments, the author decided to make a study of the respective literature. Gas-solid exchange and diffusion experiments applying radioactive nuclei as tracers are especially suitable for such evaluations because, in these experiments, the concentration of the radioactive nuclei is changed, and, in some experiments, the preparations are heated. These are just the procedures by which the anomalies of tritium radioactivity were found.[1-3] It was indeed possible to find experiments that point to a strong decrease in radioactivity of heavy nuclei (^{65}Zn, ^{63}Ni, ^{85}Sr) under certain conditions. In all these experiments, the second law of thermodynamics seems to be grossly violated. It is most striking that the radioactivity decreases several times with increasing concentration within different experiments. A further surprising statement was made: By pure formal application of the nuclear-pair hypothesis, all such deviations from normal behavior can be explained. Thus, our evaluations not only point to a strong decrease in radioactivity; they also provide further support for the nuclear-pair hypothesis for heavy nuclei.

The nuclear-pair hypothesis derived from experiments with tritium reads explicitly: Two tritium nuclei (identical nuclei with a half-integer spin) under certain conditions (in our case, embedded in small monocrystalline particles of titanium) arrange themselves in a pair with nuclear spin zero. Such a nuclear pair acts, to a certain extent, as the parent nucleus for the decay so that the radioactive decay changes, to a certain extent, into a higher, forbidden one.

Two evaluation procedures are applied that strengthen the validity of the nuclear-pair hypothesis for heavy nuclei. In the gas-solid exchange experiments and in the diffusion experiments in question, the radioactivity a is measured, and in most of these experiments, the concentration c (in the tritium experiments indicated as x) is determined theoretically, in the first case as a function of time and in the latter case as a function of distance from

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the source. The radioactivity a and the rise of radioactivity $\Delta a/\Delta c$ can thus be obtained as functions of the concentration c (first evaluation procedure). There may be some objection to the theoretical determination of the concentration c; for a definite proof, only measured values should be used. For the diffusion experiments, a second evaluation procedure was found, in which only measured values of the radioactivity a as a function of the distance d from the source are required and in which the theoretical function $c = f(d)$ is not needed.

However, it is very difficult to accept the strong decrease in λ and the validity of the nuclear-pair hypothesis as derived by our evaluations because such a high change of radioactivity has never been observed, and a theoretical explanation of the effect is completely lacking. Therefore, several straightforward experiments are proposed to prove or disprove the reduction of radioactivity and to provide further evidence for the nuclear-pair hypothesis for heavy nuclei.

II. GAS-SOLID EXCHANGE EXPERIMENTS

The work by Secco[4] and Secco et al.[5] on the exchange reaction of zinc atoms in the vapor phase labeled by the ^{65}Zn radionuclide with polycrystalline ZnO (Ref. 4) and Zn_2SiO_4 (Ref. 5) is particularly interesting. In one of the exchange curves of Ref. 5 (Fig. 3, 1000°C) the equilibrium activity for $t \rightarrow \infty$ could not be reached. To preserve the second law of thermodynamics, we are forced to assume that the radioactivity in the preparation at the end of the experiment is decreased by 30%. This experiment has the same persuasive power as the tritium heating experiments described in Refs. 1, 2, and 3.

In one of the exchange experiments of Ref. 4 (Fig. 2b, 760°C), an unexpected decrease in radioactivity was observed at $t = 39$ min. This abnormal behavior was strengthened by a second measurement. A decrease in the concentration of the ^{65}Zn atoms in the sample with further heating grossly contradicts the second law of thermodynamics. Secco was unable to interpret this apparent anomaly and has drawn explicit attention to this fact (see p. 190 in Ref. 4).

In such a gas-solid exchange experiment, the concentration of the ^{65}Zn atoms in the solid sample increases continually, resulting in increasing entropy until the ratio of the ^{65}Zn atoms to the other zinc atoms is the same in the solid and gas phases (maximum of entropy). To preserve the second law of thermodynamics, we are forced to assume that the radioactivity decreases with increasing concentration ($\Delta a/\Delta c < 0$). To begin with, we explain this extremely strange behavior by an interaction of the added ^{65}Zn nuclei with ^{65}Zn nuclei still present in the preparation, which results in a strong decrease of λ. I interpret this interaction by pair formation with $\lambda_2 \ll 0.5\,\lambda_1$ (λ_1 = decay constant of single nuclei, λ_2 = decay constant of one nucleus in a pair). However, we have to realize that it is not possible to distinguish between individual nuclei because they are identical particles; thus, it is not possible to distinguish in the pair formation process between nuclei still present in the preparation and newly added nuclei.

Furthermore, the total exchange curve in question can be evaluated by the nuclear-pair hypothesis. Figure 1 gives $a = f(c)$ and $\Delta a/\Delta c = f^x(c)$ obtained by the first evaluation procedure as explained in Sec. I. The values of a and t are taken from an enlarged photograph of Fig. 2 in Ref. 4. The values for the concentration c are taken by the same procedure from another exchange curve of Secco (Fig. 1, 760°C of Ref. 4), where, probably on account of other conditions, no decrease in radioactivity during heating occurred. The $a = f(c)$ graph is obtained by elimination of the parameter t from the $a = f^{xx}(t)$ and the $c = f^{xxx}(t)$ graphs.

We arrive at the function $\Delta a/\Delta c = f^x(c)$ by differentiation of the function $a = f(c)$. The given uncertainties in Fig. 1 are derived from the maximal experimental uncertainty of 1.5% given by Secco. Several distinct minima appear in the $\Delta a/\Delta c = f^x(c)$ graph, even with $\Delta a/\Delta c < 0$. The unit of $\Delta a/\Delta c$ in Fig. 1 was developed from the values of a and c given in Fig. 1, both in percentages by the given procedure.

The total course of the graph $\Delta a/\Delta c = f^x(c)$ can be interpreted with the nuclear-pair hypothesis, as could the $\Delta a/\Delta x = g(x)$ graph in our TiT_x experiment.[1-3] (In our

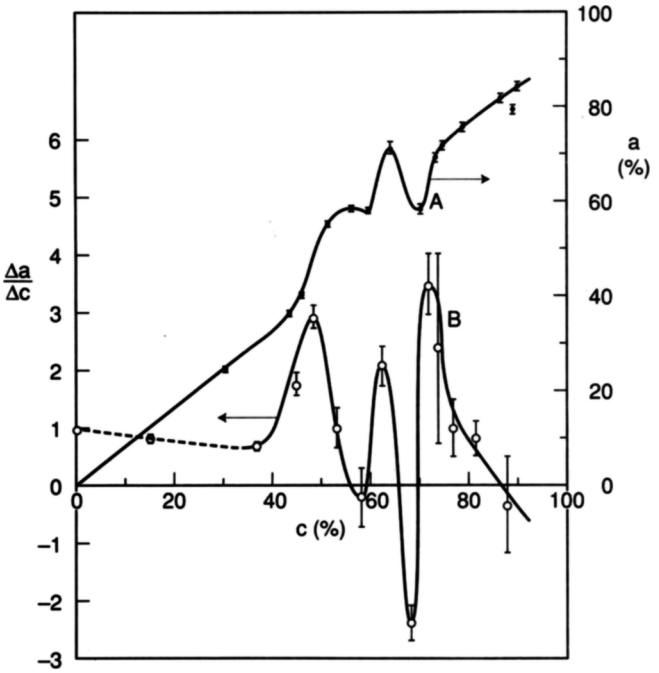

Fig. 1. Plot line A: Radioactivity a. B: Increase in radioactivity $\Delta a/\Delta c$ of ^{65}Zn as a function of concentration c during embedding in crystalline ZnO, derived from gas-solid exchange curve in Fig. 2b of Ref. 4.

tritium experiments the concentration was indicated by x.) At the minimum of $\Delta a/\Delta x$ in Fig. 3 in Ref. 1, newly added tritium nuclei resulted in a smaller increase of the radioactivity a. This was interpreted as being due to the formation of nuclear pairs with decreased λ. In our Fig. 1, this effect is even more clearly visible. There is a repeated smaller increase of the radioactivity at different increasing concentrations, even with $\Delta a/\Delta c < 0$. A detailed evaluation by the introduction of "domains where pair formation takes place" and the connection with concentration is given in Sec. III. One has to realize that in the gas-solid exchange experiment of Secco, the repeated nuclear pairing at different concentrations cannot be seen at the beginning of the experiment. This information is apparently lost by the fast increase of concentration in this region of c.

There are several other exchange curves in Secco's paper[4] that provide equivalent results. The most striking is that of his Fig. 1 with $p_{Zn} = 0.11$ atm and $S = 0.5$. Again several minima of the $\Delta a/\Delta c = f^x(c)$ graph are obtained. However, the evaluation by the first evaluation procedure has less persuasive power than the evaluation of the experiment reported earlier because the measurements in question were only done once, and the "normal" exchange curve given by Secco was obviously obtained taking mean values. The real exchange curve is probably steeper after $t = 30$ min. On the other hand—if we trust Secco's specification concerning the accuracy of his measuring procedure and the validity of the second law of thermodynamics—the repeated decrease of a at $t = 39$, 65, and 79 min again indicates several decreases in the ^{65}Zn radioactivity with increasing concentration ($\Delta a/\Delta c < 0$). For example, at $t = 39$ min, a is decreased by ~10% with respect to the preceding measurement point at $t = 31$ min and by ~19% with respect to the value of the expected a at $t = 39$ min, if we assume continual increase of concentration with time. Both deviations are very much larger than the maximum experimental error of 1.5%.

III. DIFFUSION EXPERIMENTS

Another category of experiments relates to investigations into diffusion in solids using radioactive tracers. I have made evaluations based on measurements taken from a publication by Atkinson and Taylor[6] (Fig. 2, run 10 therein) on the diffusion of nickel in NiO single crystals using ^{63}Ni as a tracer and from a paper by Torstenfelt et al.[7] (Fig. 5 therein) on the diffusion of ^{85}Sr in compacted bentonite clay. In the latter experiment the ^{85}Sr atoms are chemically sorbed on the small crystalline clay particles. It is most interesting that with the preparation method of Atkinson and Taylor, small particles are obtained under certain conditions (gas pressure and rate of sputtering), which was also the case with the preparation of the titanium samples applied by the author in the tritium experiments.[1-3] The activities are determined by the same manner as in the gas-solid exchange experiments of Secco. All the graphs $\Delta a/\Delta c = f(c)$ derived by the first evaluation procedure from the aforementioned diffusion experiments show a trend equivalent to that of the curves obtained from the gas-solid exchange experiments, namely, periodic variations of $\Delta a/\Delta c$ with increasing c even with a decrease in the activity a at increasing concentration c ($\Delta a/\Delta c < 0$). The most important objection to this evaluation procedure is the fact that the concentration c as a function of the distance d is only determined theoretically.

However, there is another quite different and independent method of evaluation, indicated as the second evaluation procedure, where only measured values of the radioactivity a as a function of the distance d from the source are used, and the theoretical curve $c = f(d)$ is not needed. Figure 2 shows the penetration profile of run 10 taken from Fig. 2 of Atkinson and Taylor's paper,[6] but with a linear scale for the activity a. Near the source, the activity a oscillates very strangely with increasing distance d. This peculiar behavior is somewhat camouflaged in Fig. 2 of Ref. 6 by the logarithmic scale of the activity a. The diffusion coefficient calculated in Ref. 6 is actually obtained from the part of the profile with $d >$ 0.85 μm, where the concentration is low. They calculated $D = 1.13 \times 10^{-15}$ cm^2/s.

The penetration profile as shown with the linear scale for a is such that it is completely impossible to get a reasonable diffusion coefficient with the conventional evaluation

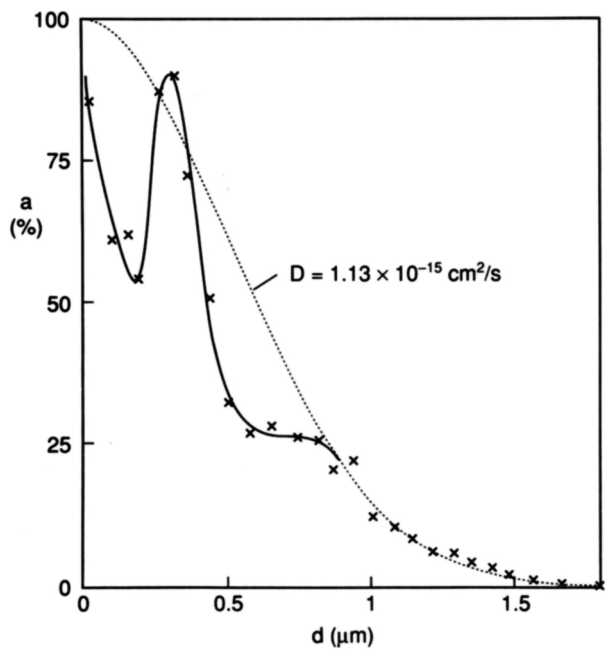

Fig. 2. Penetration profile of ^{63}Ni diffusion in NiO single crystal of Ref. 6, Fig. 2, run 10. Radioactivity = a, distance = d, plotted on a linear scale of a.

method for $d < 0.85$ μm, which is just the region where ~70% of the variation of a takes place. For example, according to Fig. 2, the ^{63}Ni nuclei diffuse between $d = 0.17$ μm and $d = 0.295$ μm against an increasing concentration, contradicting the laws of diffusion. To preserve this law we again have to assume that the radioactivity decreases with increasing concentration ($\Delta a/\Delta c < 0$). Our nuclear-pair hypothesis can solve this puzzle; it provides a means by which a diffusion coefficient can be calculated from the strange graph $a = f(d)$ presented in Fig. 2 for $d < 0.85$ μm and thus proves that normal diffusion exists in this region. If we measure the radioactivity of our samples beginning with low concentration ($d \approx 2$ μm) and proceed to smaller distances d, we have a continually increasing concentration of the radionuclide in our samples, and we do, in principle, obtain the same as in our TiT_x experiment.[1,3]

To arrive at an explanation of the strange diffusion profile of Fig. 2, we assume that our sample is divided in domains where pair formation takes place. These domains were thus successively filled, with 1,2,3,..., ^{63}Ni nuclei. If all domains are filled with a certain number ζ, we attribute the occupation number ζ to the respective concentration c corresponding with a distance d. Coming from low concentration, one has, as stated earlier, normal diffusion: The domains where pair formations take place are filled with one ^{63}Ni nucleus. No nuclear pairing can thus occur, and we have normal diffusion. At $d = 0.85$ μm, there is a sudden deviation from the normal diffusion profile to smaller values of a, and $\Delta a/\Delta c$ is decreased to nearly zero in the region between $d = 0.85$ and 0.55 μm. According to our assumption, at $d = 0.85$ μm all the domains where pair formation takes place are just filled with one ^{63}Ni nucleus, and any further added nuclei form pairs, resulting in a smaller increase of a. We have thus to attribute the occupation number $\zeta = 1$ to the sample with $d = 0.85$ μm.

After $d = 0.55$ μm we see a sharp increase in the activity. This can be explained by the assumption that at $d = 0.55$ μm all domains where pair formation takes place are just filled with two ^{63}Ni nuclei, and the newly added single nuclei have normal λ. The relating sample therefore gets the occupation number $\zeta = 2$. The sample taken at $d = 0.55$ μm corresponds to the minimum of $\Delta a/\Delta x$ in our TiT_x experiment, after which $\Delta a/\Delta x$ increases (Fig. 3 of Ref. 1). At $d = 0.295$ μm we have a maximum of a and thus a sharp change of $\Delta a/\Delta c$ from positive to negative values. Nuclear pairing starts again even with $\Delta a/\Delta c < 0$, thus with a sharp decrease of λ. We assign the related sample the occupation number $\zeta = 3$. The decrease of a between $d = 0.295$ and 0.17 μm with decreasing distance d from the source, thus with increasing concentration of the radionuclide, strengthens our hypothesis of the decrease of the radioactivity of a heavy radionuclide ($\Delta a/\Delta c < 0$). At $d = 0.17$ μm we have a minimum of a and $\Delta a/\Delta c$ changes from negative values (pair formation) to positive ones, which means that new single ^{63}Ni nuclei are added that exhibit "normal" radioactivity. Thus, we have to give this sample the occupation number $\zeta = 4$.

By this procedure we have occupation numbers ζ as a function of the distance d. We thus have a concentration $\zeta \equiv c$ as a function of the distance d and can calculate a diffusion coefficient D by applying the solution of the one-dimensional diffusion equation:

$$C\,(\%) = 100 \exp\left(-\frac{d^2}{4tD}\right) ,$$

$$D = \frac{d_\mu^2 - d_\nu^2}{4t(\ln \zeta_\nu - \ln \zeta_\mu)}$$

with

$$t = 1.2 \times 10^6 \text{ s} .$$

If we take the most distant samples $d = 0.85$ μm, $\zeta = 1$ and $d = 0.17$ μm, $\zeta = 4$, by this procedure we get a diffusion coefficient $D = 1.04 \times 10^{-15}$ cm^2/s, which is almost equal to the value calculated by Atkinson and Taylor[6] ($D = 1.13 \times 10^{-15}$ cm^2/s) from their penetration profile, which they, in fact, only calculated from the profile with $d > 0.85$ μm, where the concentration is so low that no nuclear pairing can occur.

In Table I the results of this evaluation are presented. A general criterion to find the distance d for the different occupation numbers is also given in the table. With $\Delta^2 a/\Delta c^2 < 0$ ($\Delta a/\Delta c$ decreases), one has uneven occupation numbers, and with $\Delta^2 a/\Delta c^2 > 0$ ($\Delta a/\Delta c$ increases), one has even occupation numbers. If one starts with the first $\Delta^2 a/\Delta c^2 < 0$ and gives the related sample the occupation number 1, the occupation numbers follow automatically.

I applied our second evaluation procedure to further analyze the penetration profile measured by Torstenfelt

TABLE I

Occupation Numbers ζ Determined from the Experimental $a = f(d)$ Curve of the ^{63}Ni Diffusion Experiment of Ref. 6, According to the Second Evaluation Procedure

Determination of the Diffusion Coefficient for the Most Distant Occupation Numbers

d (10^{-4} cm)	$\Delta^2 a/\Delta c^2$	ζ	ζ	ζ	D (10^{-15} cm^2/s)
0.85	<0	Uneven	1 ↑[a]	1 ↑	
0.55	>0	Even	2 ↑		
0.295	<0	Uneven	3 ↑		1.04
0.17	>0	Even	4	4	

[a]The arrows indicate the direction of the diffusion.

et al.[7] of the diffusion of ^{85}Sr in bentonite clay (Fig. 5 of Ref. 7) on the left and right. Bentonite consists of small clay particles of ~100 Å in size that are stacked to form aggregates of micron size. When water is added, it enters between the individual small clay particles, and an exchange between cations in the water and cations in the small clay particles takes place. In this manner, ^{85}Sr cations are built into the small clay particles.

Figure 3 gives the measured activities $a = f(d)$; the a values of the ^{137}Cs diffusion, done in the same experiment, are removed for more clarity. After an initial sharp decrease in a with increasing d, one observes wild fluctuations of a for both sides of the experiment (the "sides" concern Fig. 3). It was impossible for the authors of Ref. 7 to calculate a diffusion coefficient from these penetration profiles (see p. 298 of Ref. 7).

The strong scatter of a cannot be caused by statistical or other experimental uncertainties. If we assume experimental uncertainties as the cause of the scatter, we should be able to determine a diffusion coefficient for both sides of the experiment with $d > 2.4$ mm by a least-squares fit. This turns out to be completely impossible. Thus, the assumption that the scatter of the measured values in Fig. 3 is caused by experimental errors leads to a strong contradiction.

We again apply our second evaluation procedure, which is based on the nuclear-pair hypothesis. Tables II and III give the results. Because of the rather coarse setup of the experiment, the most distant samples with definite occupation number ζ are taken for the calculation of the diffusion coefficient. One gets $D = 6.3 \times 10^{-13}$ m^2/s for the right side of the experiment and $D = 5.7 \times 10^{-13}$ m/s^2 for the left side of the experiment. If one shifts the maximum of the $a = f(d)$ graph 0.2 mm to the right, as suggested by the maximum of the ^{137}Cs penetration profile in Fig. 5 of Ref. 7, one gets $D = 6.0 \times 10^{-13}$ m^2/s for both sides of the experiment. At the left side we obviously have "normal" diffusion between $d = 5.2$ and 5.6 mm, and we can calculate the normal diffusion coefficient applying the conventional method. The result, $D = 8.3 \times 10^{-13}$ m^2/s, is in sufficient agreement with the diffusion coefficient determined by the second evaluation procedure, taking the rather course determination of the normal diffusion coefficient into account. Again in this experiment, we observe a decrease of radioactivity with increasing concentration several times.

It can be said that the course of $a = f(d)$ between 2.4 and 5.6 mm is accidentally caused by differences in the sample thickness. However, then the $a = f(d)$ graph for the ^{137}Cs diffusion would have to show the same scatter, and this certainly is not the case. Disturbance by the walls of the diffusion chamber cannot be the cause of the fluctuations either, because then they would also have had to have been observed in all the other diffusion measurements of Ref. 7.

The term "domain where pair formation takes place" is introduced ad hoc to arrive at a preliminary explanation of the periodicity of $a = f(c)$ and $\Delta a/\Delta c = f^x(c)$. It may be transformed into concentration or into distance between the nuclei. For a precise determination of $a = f(c)$

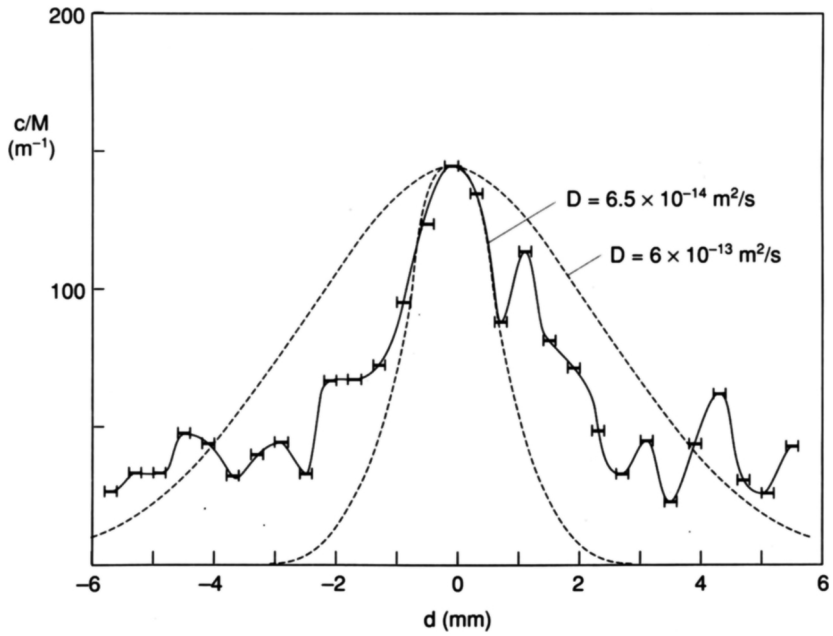

Fig. 3. Penetration profile of ^{85}Sr diffusion in compacted bentonite clay of Fig. 5 of Ref. 7, the right and left sides of the experiment. Theoretical profiles are prepared with $D = 6 \times 10^{-13}$ m^2/s and $D = 6.5 \times 10^{-14}$ m^2/s. Radioactivity a is in counts per minute (c/M).

TABLE II

Occupation Numbers ζ Determined from the Experimental $a = f(d)$ Curve of the ^{85}Sr Diffusion Experiment,[7] According to the Second Evaluation Procedure, Right Side of the Experiment

Determination of the Diffusion Coefficient for the Most Distant Extrema and with Source ($d = 0$) Shifted 0.2 mm to the Right

d (10^{-3} m)	Extrema	$\Delta^2 a/\Delta c^2$	ζ	ζ	ζ	D (10^{-13} m^2/s)	Source Shifted 0.2 mm to the Right		
							d (10^{-3} m)	ζ	D (10^{-13} m^2/s)
5.6	Max	<0	Uneven	1 ↑[a]	1 ↑		5.4	1 ↑	
5.2	Min	>0	Even	2 ↑					
4.4	Max	<0	Uneven	3 ↑		6.3			6.0
3.6	Min	>0	Even	4 ↑					
3.2	Max	<0	Uneven	5 ↑					
2.8	Min	>0	Even	6	6		2.6	6	

[a]The arrows indicate the direction of the diffusion.

TABLE III

Occupation Numbers ζ Determined from the Experimental $a = f(d)$ Curve of the ^{85}Sr Diffusion Experiment,[7] According to the Second Evaluation Procedure, Left Side of Experiment

Determination of the Diffusion Coefficient for the Most Distant Extrema and with Source ($d = 0$) Shifted 0.2 mm to the Right

d (10^{-3} m)	Extrema	$\Delta^2 a/\Delta c^2$	ζ	ζ	ζ	D (10^{-13} m^2/s)	Source Shifted 0.2 mm to the Right		
							d (10^{-3} m)	ζ	D (10^{-13} m^2/s)
5.2	Max	<0	Uneven	1 ↑[a]	1 ↑		5.4	1 ↑	
4.8	Min	>0	Even	2 ↑					
4.4	Max	<0	Uneven	3 ↑		5.7			6.0
3.6	Min	>0	Even	4 ↑					
2.8	Max	<0	Uneven	5 ↑					
2.4	Min	>0	Even	6	6		2.6	6	

[a]The arrows indicate the direction of the diffusion.

and $\Delta a/\Delta c = f^x(c)$, carefully designed and performed experiments are needed. The information contained in Figs. 1, 2, and 3—which, of course, is rather limited—point to a kind of sinusoidal shape of the periodic functions. The accurate shape of these functions may have a bearing on the theory of the effect.

IV. DISCUSSIONS AND PROPOSALS FOR FURTHER EXPERIMENTS

From the evaluations described in this paper, two preliminary conclusions are derived:

1. the radioactivity of heavy nuclei decreases strongly

2. the nuclear-pair hypothesis may serve as a preliminary explanation of this extremely strange effect.

These preliminary conclusions are supported by the following observations:

1. It has been observed several times that the radioactivity of a preparation decreases with increasing concentration. This extremely strange behavior can be explained by an interaction between the nuclei, and I interpret it preliminarily by pair formation with strongly decreased λ ($\lambda_2 \ll 0.5\lambda_1$; λ_1 = decay constant of unpaired nuclei, and λ_2 = decay constant of one nucleus in a pair).

2. In some experiments, we see, beginning with low concentration c and slowly increasing c, a periodic increase and decrease of a and, in other experiments, of $\Delta a/\Delta c$. I preliminarily interpret this observation as being due to successive formation of nuclear pairs at certain concentrations or distances between the nuclei.

It is most important that observations 1 and 2 are supported by two essentially different experiments:

1. In the gas-solid exchange experiment of Secco,[4] the concentration is slowly increased with increasing time in one sample at a distinct place.

2. In the diffusion experiments,[6,7] one obtains different samples with increasing concentration at a distinct time.

The fact that the aforementioned observations are made in both essentially different experiments supports the conclusions. That a decrease in the radioactivity of ^{65}Zn occurs is furthermore supported by the heating of a Zn_2SiO_4 sample at 1000°C, where the ^{65}Zn radioactivity was decreased by 30% at the end of the experiment (Ref. 5, Fig. 3, 1000°C).

However, the author realizes that these statements are very revolutionary because they are in conflict with longstanding accepted nuclear theories. For this reason, it is first and foremost necessary that further experimental evidence of the striking phenomena be produced. In the following sections, several experiments are proposed.

IV.A. Experiment 1

Gas-solid exchange reactions described by Secco[4] and Secco et al.[5] are suitable for the proof and further exploration of the effect. Figure 4 shows the proposal for an experimental setup. The ZnO or Zn_2SiO_4 (preparation 1) and the Zn + ^{65}Zn (preparation 2) delivering the ^{65}Zn to the preparation 1 by gas-solid exchange are separated by

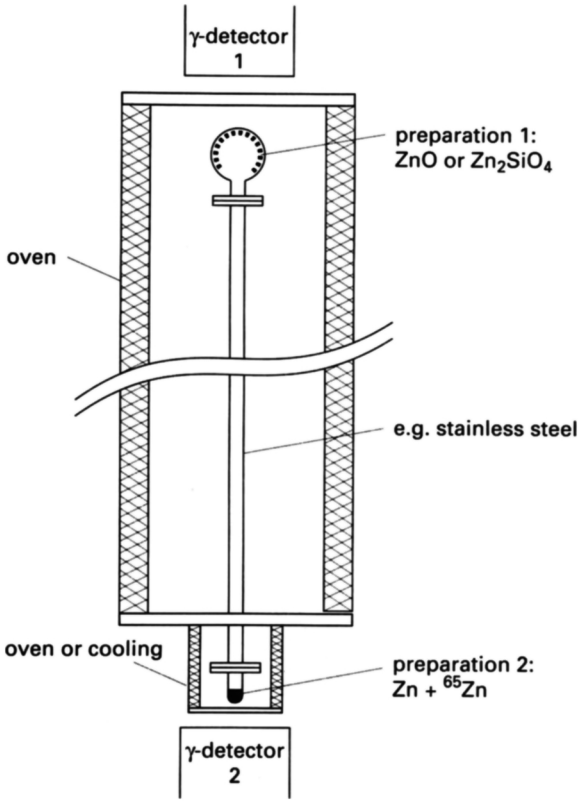

Fig. 4. Proposal for experimental arrangement for the determination of the function $a = f(c)_T$ with ^{65}Zn. Preparation 2 delivers ^{65}Zn to preparation 1 by gas-solid exchange reaction. The two detectors monitor continuously the respective activities. Here, a = radioactivity, c = concentration, and T = temperature.

sufficient distance that an independent determination of the radioactivity in preparation 1 and the ^{65}Zn-labeled preparation 2 is possible. In this experiment a determination of $a = f(c)_T$ in preparation 1 is possible beginning with low concentrations.

The ZnO or Zn_2SiO_4 powders of the same type used in the experiments of Refs. 4 and 5 can be applied. From our tritium experiments we arrived at the suspicion that monocrystallinity may be a necessary condition for the decrease of radioactivity.[2,3] Polycrystalline samples may consist of monocrystalline or nonmonocrystalline particles. The condition of crystallinity has to be investigated. Furthermore, ZnO samples can easily be prepared by gas evaporation of zinc followed by oxidation. This results in preparations with properties equivalent to those used in our tritium experiments (monocrystallinity and small size).

The major problem in experimentation, besides a suitable choice of solid preparation, is the choice of concentration of the radionuclide. It is a pity that most authors do not give this figure in their papers, but sometimes it is possible to make an estimation from their specifications.

IV.B. Experiment 2

The diffusion experiment of Atkinson and Taylor[6] also suggests a straightforward experiment for the proof of our hypothesis. This experiment[6] consists of two parts:

1. By diffusion of the ^{63}Ni in the NiO single crystal, a decreasing concentration of the ^{63}Ni as a function of the distance d from the surface is obtained. It cannot be determined from our evaluation whether the radioactivity has already decreased in the single crystal. The diffusion experiment just provides the preparation of samples with varying concentrations.

2. Samples containing the ^{63}Ni nuclei with varying concentration are then obtained by radiofrequency sputtering.

The following experiment is suggested: Using a suitable evaporator (e.g., as used in our tritium experiments and described in Ref. 8), a metal containing a radionuclide is evaporated in argon of, for example, 1 cm mercury pressure (see Refs. 1, 2, and 3). Zinc-65 is a suitable choice when we evaporate zinc. The high energy gamma radiation makes it possible to determine the radioactivity easily before and after evaporation, e.g., in a glass ball. Also heating the preparation may be applied. The decrease in radioactivity and the nuclear-pair hypothesis can be tested by applying increasing concentrations of the radionuclide in succeeding experiments.

IV.C. Experiment 3

The Torstenfelt et al.[7] experiment concerning the diffusion of ^{85}Sr in bentonite clay with binding of the radionuclide on the crystalline clay particles (exchange with Na-ions) also suggests straightforward experimentation. We consider the sample 2.8 mm from the source on the right side of the experiment (or 2.4 mm on the left side of the experiment) with occupation number $\zeta = 6$ (Fig. 3). During the experiment the concentration of the radionuclide chemisorbed on the clay particles increases continuously from $\zeta = 0$ to 6, and, according to our hypothesis, we obtain increasing and decreasing radioactivities. This suggests that we increase the concentration of the radionuclide continuously in an aqueous suspension or in a sediment of the clay particles with continuous detection of the radioactivity. Intermittent separation of the solid from the aqueous phase should be performed, and the radioactivity in both phases should be determined. It is recommended that the experiment begin with the same components (Wyoming bentonite MX-80, ^{85}Sr, synthetic groundwater) as those used by the authors of Ref. 7.

IV.D. Experiment 4

The experiments in which we continuously increase the concentration of the radionuclide are especially suitable to prove or disprove the nuclear-pair hypothesis. A further interesting possibility is the preparation of a suitable radionuclide in situ by irradiation of a finely divided preparation, e.g., in a nuclear reactor. Such a preparation may be preferably prepared by gas evaporation, possibly followed by oxidation. Elevated temperatures should be applied during or after irradiation.

V. CONCLUDING REMARKS

It is highly probable that a hidden necessary condition for the decrease of λ may exist. As I have pointed out in Refs. 2 and 3, monocrystallinity may be such a condition for the decrease in tritium radioactivity and also for the cold fusion phenomenon.

The evaluation of our gas-solid exchange and diffusion experiments does not provide the possibility to distinguish between an interaction of this type of tritium experiment,[1-3] in which the normal radioactivity can be recovered, and a permanent fusion. The latter occurs in cold deuterium-deuterium fusion[9] and in fusion processes claimed by Sundaresan and Bockris[10] and by Singh et al.[11] By solving the preparations in which a decrease in radioactivity is stated in a suitable acid or base, one can simply distinguish between these two possibilities. This may be also a method of regaining the original radioactivity.

In all the experiments evaluated to date, the radioactive nuclei were fermions. It should be investigated whether the effect also occurs with bosons. For example, ^{60}Co is a suitable radionuclide for relating experiments.

All of the experiments evaluated in this paper have concerned beta decay and electron capture. It would be interesting to ascertain whether the decrease in radioactivity also concerns alpha decay.

Nuclear pairing could be proved by nuclear magnetic resonance (NMR) experiments. It may be interesting that V. D. Lugt,[12] who investigated finely divided copper samples prepared by the author by gas evaporation, received neither an NMR signal of ^{63}Cu nor of ^{65}Cu at room temperature nor at the temperatures of liquid helium. The absence of the NMR signal could simply be caused by the small dimensions of the copper particles. By increasing the argon pressure, the monocrystallinity of the particles was lost; however, the size of the particles was also increased. Then an NMR signal was observed. The sensitivity of NMR measurements is today many orders of magnitude higher than it was during the experiments of V. D. Lugt, and it would certainly be interesting to do related experiments.

As I have pointed out, the decrease in radioactivity and the explanation using the nuclear-pair hypothesis are in conflict with longstanding accepted nuclear theories: It is very difficult to imagine that nuclei with dimensions of $\sim 10^{-12}$ cm interact with other nuclei separated by

distances of several 10^{-8} cm. With regard to this, I should like to remind the reader that there is a well-known example for pair formation of elementary particles at a long distance: the Cooper pairs in superconductivity. A new entity is formed; the mass is experimentally determined as $2m_e$, the electrical charge as $2e$, and the spin as zero. However, if an analogous pair formation were to exist between nuclei, this could explain the experiments of V. D. Lugt[12] but would not be sufficient for a theoretical explanation of the decrease of radioactivity. It is quite incomprehensible that the weak interaction occurring in a nucleus could be influenced by another nucleus situated at a distance of several nanometres.

It is my hope that this paper will stimulate further experimental research on these striking phenomena.

ACKNOWLEDGMENTS

The author would like to thank H. B. G. Casimir, H. L. Hagedoorn, D. Lenstra, M. H. Steenland, B. J. Verhaar, and M. J. A. de Voigt for their interest in this work and for valuable discussions.

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