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Emission of DD-Fusion Neutrons from a Massive Palladium Cylinder during Electrolytic Infusion of Deuterons into the Metal

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A weak emission of fast neutrons during the long-duration loading of a massive palladium cylinder with deuterons is observed. The experimental results are discussed in the frame of a plasma-like model of nuclear fusion in condensed matter, resulting in fusion rates per interacting pair of $\lambda_{dd}^{Pl} = (1.05 \pm 0.15) \cdot 10^{-44}\,s^{-1}$. *This effect is also discussed relative to the behaviour of an other, similar cathode, which was charged simultaneously with the one described in this paper.*

Während der langandauernden Beladung eines massiven Palladium-Zylinders mit Deuteronen wird eine schwache Emission schneller Neutronen beobachtet. Die experimentellen Ergebnisse werden im Rahmen eines plasma-änlichen Modells der Kernfusion in kondensierter Materie diskutiert, woraus eine Plasmafusionsrate von $\lambda_{dd}^{Pl} = (1.05 + 0.15) \cdot 10^{-44}\,s^{-1}$ *resultiert. Dieser Effekt wird auch relativ zum Verhalten einer anderen, ähnlichen Kathode diskutiert, welche gleichzeitig mit der in der vorliegenden Arbeit beschriebenen beladen wurde.*

Keywords

deuterium; deuterons; electrolysis; electrolytic cells; fast neutrons; fusion yield; nuclear physics; nuclear reactions; palladium; plasma

1. Introduction

Recently, in several experiments there was found an evidence for a weak neutron production by supposed dd-fusion processes in condensed matter [1–3]. At the TU Dresden after first confirming experiments [3] a limited programme on systematic studies of this phenomenon was carried out.

The experimental method used for this purpose was described elsewhere [4]. In this paper also the investigation of the background and results of measurements using H_2O instead of D_2O are presented.

For physical discussions a simple plasma model describing the time-dependence of assumed dd-fusion processes was proposed in [5]. We refer to this two papers [4, 5] for all the details of both experimental methods and physical model applied. The present paper describes results of a long term experiment using the massive palladium cylinder Z2. This electrode was under observation simultaneously with two other electrodes: One of them is a similar cylinder Z1, the behaviour of which is presented in the paper [6]. The second is a massive slab AH1, which will be published elsewhere.

2. Experiment

The experimental cycle series 3 described here had a total duration of 786 hours, but the electrolytic charging of the cylinder Z2 was started only 50 hours later than that of cylinder Z1 [6]. But in opposite to Z1 it remained in the experiment until its end, this means for 736 hours, totally.

As described in [4], all measurements of neutron emission from cell with Z2 were carried out strictly relative to the other electrodes (cells in front of the proton recoil spectrometer PRS) and to the background (empty position) in a sequence of single one-hour measurements. Therefore, each cell was under observation near to the PRS only for a part time of the total duration of the cycle. For Z2 a total amount of 142 measurements was carried out, during this time the background was measured 146 times.

The unloaded high purity Palladium cylinder Z2 had the following dimensions: diameter $d = 22.6$ mm, length $l = 20.2$ mm, unloaded weight $m_{Pd} = 92.691$ g.

The other experimental conditions are identical or similar to those, which were reported for Z1 [6]: electrolyte 3M solution of LiOD in high purity D_2O, current I = 4A a.s.o. The proton recoil energy PRE measured with the liquid scintillation detector was recorded both in 512 channels and in more sensitive broad channel groups (ranges):

channel range 2: PRE 1.4–2.9 MeV
channel range 3: PRE 1.9–2.9 MeV
channel range 4: PRE 2.9–4.1 MeV
channel range 5: PRE 4.1–6.5 MeV

The best effect to background ratio for dd-neutrons is expected for channel range 3.

The method used is very sensitive and allows to eliminate the influence of possible slow shifts of both the electronics and cosmic background, but it does not allow for the observation of short bursts, as all the effects are averaged over one hour, at least.

After the experimental cycle finished, the deuterium content in Z2 was determined by weighing: Z2 was charged with $m_D = 1.424$ g deuterium, corresponding to a D:Pd atomic ratio of 0.812. This number was obtained comparing the weight of the dry charged cathod immediately after the end of the experimental cycle with the weights of the pure metallic sample both before charging and after its outgassing at high temperature in a vacuum stove.

3. Experimental results

In Fig. 1 the distribution of experimental counting rates in channel range 3 with cell Z2 (right side) and empty position (left side) for different running time intervals is shown. As it was observed for Z1 [6] also in the present case for the time interval 50 ... 200 h this difference is negative. This again is a consequence of the shadow effect, discussed in [4], which results in an effective reduction of the channel range 3 background line by $(-1.4 \pm 1.0)\,h^{-1}$. Even if this effect is not taken into account, the counting rate differences for the next two intervals are definitely positive and there is also a positive difference for the very long total time interval between 50 and 740 hours. In Tab. 1 the uncorrected and corrected counting rate differences in the channel range 3 are summarized. The corrected difference counting rate starts with zero within 1σ and increases to almost 3σ in the third time interval. Of course, there are smaller time intervals in which counting rate differences are higher.

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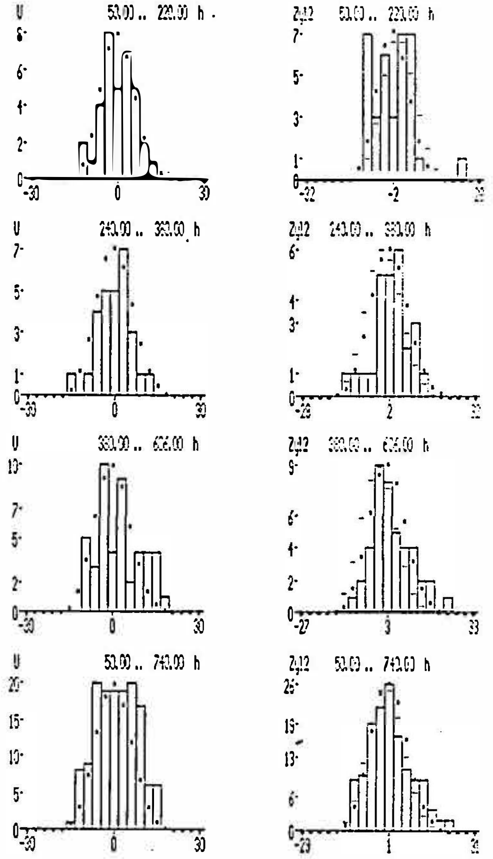

Fig. 1. Distributions of counting rate differences for empty position (left side) and with cell Z2 (right side) in comparison with $\varkappa^2$-minimum background line obtained from background measurements; this distributions are presented for different time intervals as indicated in the figure; additionally the shape of Poisson distributions is indicated by points

For instance, in the interval 380 ... 400 h the corresponding uncorrected and corrected numbers are $(4.1 \pm 0.6)\,\mathrm{h}^{-1}$ and $(5.5 \pm 1.1)\,\mathrm{h}^{-1}$, respectively. The largest single uncorrected effect counting rates are in the order of $(20 \pm 5)\,\mathrm{h}^{-1}$. As a matter of fact there are definite positive effects in partial time intervals of a few days duration.

Tab. 1. Averaged effect (cell Z2) minus background counting rate differences for channel range 3 within different time intervals

time interval [h]	measured difference $[\mathrm{h}^{-1}]$	corrected difference $[\mathrm{h}^{-1}]$
50 ... 220	-2.3 ± 1.2	-0.9 ± 1.6
240 ... 380	$+2.0 \pm 1.2$	$+3.4 \pm 1.6$
380 ... 606	$+3.5 \pm 1.5$	$+4.9 \pm 1.8$
50 ... 740	$+1.1 \pm 0.7$	$+2.5 \pm 1.2$

To get information about the time dependence of supposed dd-reaction processes, the counting rate differences are averaged over longer time intervals, as shown in Fig. 2, where the averaging interval is taken equal to 10, 40 and 60 hours of the running time. In this way it becomes evident, that there is a slow increase of the average counting rate difference with a broad maximum around $t = 400$ h. This is very similar to the behaviour which was observed for Z1 [6], but shifted in time just for about 50 hours, i.e. the starting time delay of Z2. Again we come to the conclusion, that after averaging over fluctuations and short-time peaks and valleys, a small but for a long time stable positive effect over several hundred hours is observed. The dashed lines in this and the following figures represent the effective background line.

In Fig. 3 the s.c. integrated counting rate differences [4] for the different channel ranges of interest is shown. In comparison with the corrected background level in both channel ranges 2 and 3 the integrated difference is definitely positive. The increase of the integrated counting rate difference between 200 h and 400 h in channel ranges 2 and 3 is not observed in the channel ranges 4 and 5.

The cumulative effect counts are presented in Fig. 4 for the channel ranges 2, 3, 4 and 5. There is a definite deviation from the declining effective background line (dashed line) for channel groups 2 and 3. For channel range 4 it is nearly zero or at least much smaller (multiple dd-neutron emission could give a small contribution in this channel range). For channel range 5 the cumulative effect is zero within 1σ.

For a correct interpretation of the cumulative effect one must take into account, that as mentioned above within 736 h of charging the electrode was under observation for 142 h only. Therefore, the cumulative counting rate has to be increased by the factor 5.18, to get counting rates, which are equivalent to the whole duration of the experimental cycle.

If dd-reaction rates are determined, additionally a factor of 33.3 has to be multiplied for accounting the 3% detection efficiency in channel range 3 [4]. (This includes both the neutron detector efficiency for dd-neutrons and the geometrical factor.) Therefore, the cumulative number of counts shown in Fig. 4 for channel range 3 corresponds in average to about $(5.3 \pm 0.6) \cdot 10^4$ dd-reaction events within 736 hours.

This is somewhat less than it was observed for Z1 [6], but comparable within the uncertainty limits.

Finally, let us look at the difference of counting rates for cells with Z1 and Z2, shown in Fig. 5. From this presentation it is evident, that the different cells are not equally active at the same time. In the time interval 240 ... 380 h cell Z1 shows a higher activity than cell Z2, whereas in the time interval 380 ... 606 h the cell Z2 in average is more active. Thus, common external sources of radiation which could induce the observed effects, are very unlikely. Also, in this way a hypothetic wrong measurement of the background as a possible origin of false positive effects for both Z1 and Z2 can be excluded.

Internal processes depending on the charging of the electrodes seem to provide the more plausible explanation of the observed effects.

4. Discussion

The plasma model described in [5] provides some explanation for the gross time-structure of effects observed experimentally.

From the average counting rate differences presented in Fig. 2 the average dd-reaction rates $\dot{N}^{\mathrm{dd}}$ $[\mathrm{s}^{-1}]$ can be obtained taking into account the registration efficiency for dd-neutrons.

Following the model, the time-structure of the average reaction rate is given by

$$\dot{N}^{\mathrm{dd}} = (n_{\mathrm{D}}^{\max})^2 \frac{\pi d^2\, 1}{4} \left[1 - \exp\left(-\frac{t}{t_L}\right)\right]^2 \exp\left(-\frac{t}{t_L}\right) \varkappa \lambda_{\mathrm{dd}}^{\mathrm{Pl}}$$

$$\sim \left[1 - \exp\left(-\frac{t}{t_L}\right)\right]^2 \exp\left(-\frac{t}{t_L}\right), \qquad (1)$$

with the loading time constant t_L

$$t_L = \frac{\mathrm{d} n_{\mathrm{D}}^{\max}}{4 S_0}, \qquad (2)$$

the part of mobile particles $\varkappa$

$$\varkappa = \exp\left(-\frac{V_0}{kT}\right) \qquad (3)$$

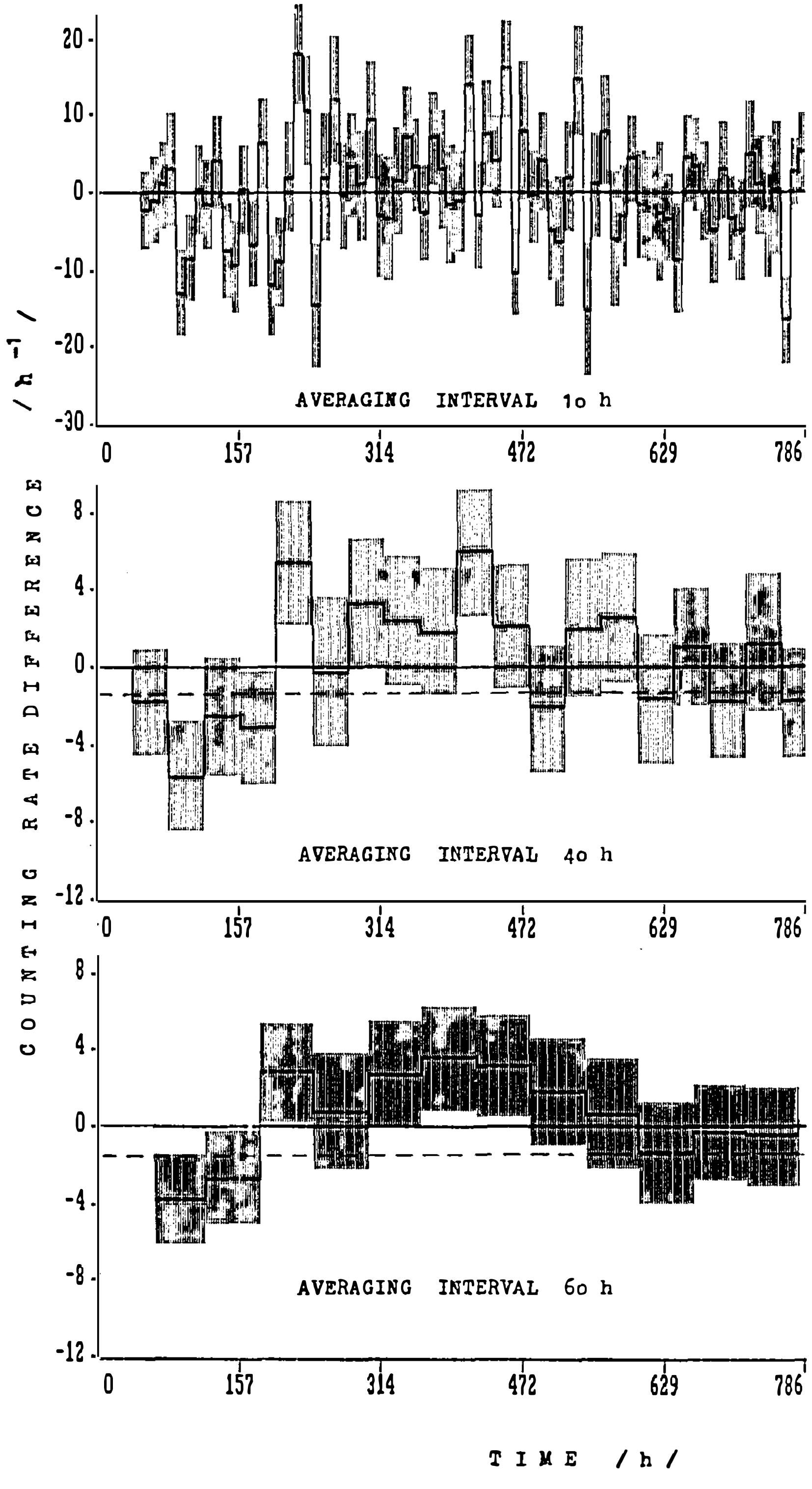

Fig. 2. Counting rate differences averaged over $\Delta t = 10$ h (upper part), 40 h (middle) and 60 h (lower part) as function of running time. Dashed line — effective background level

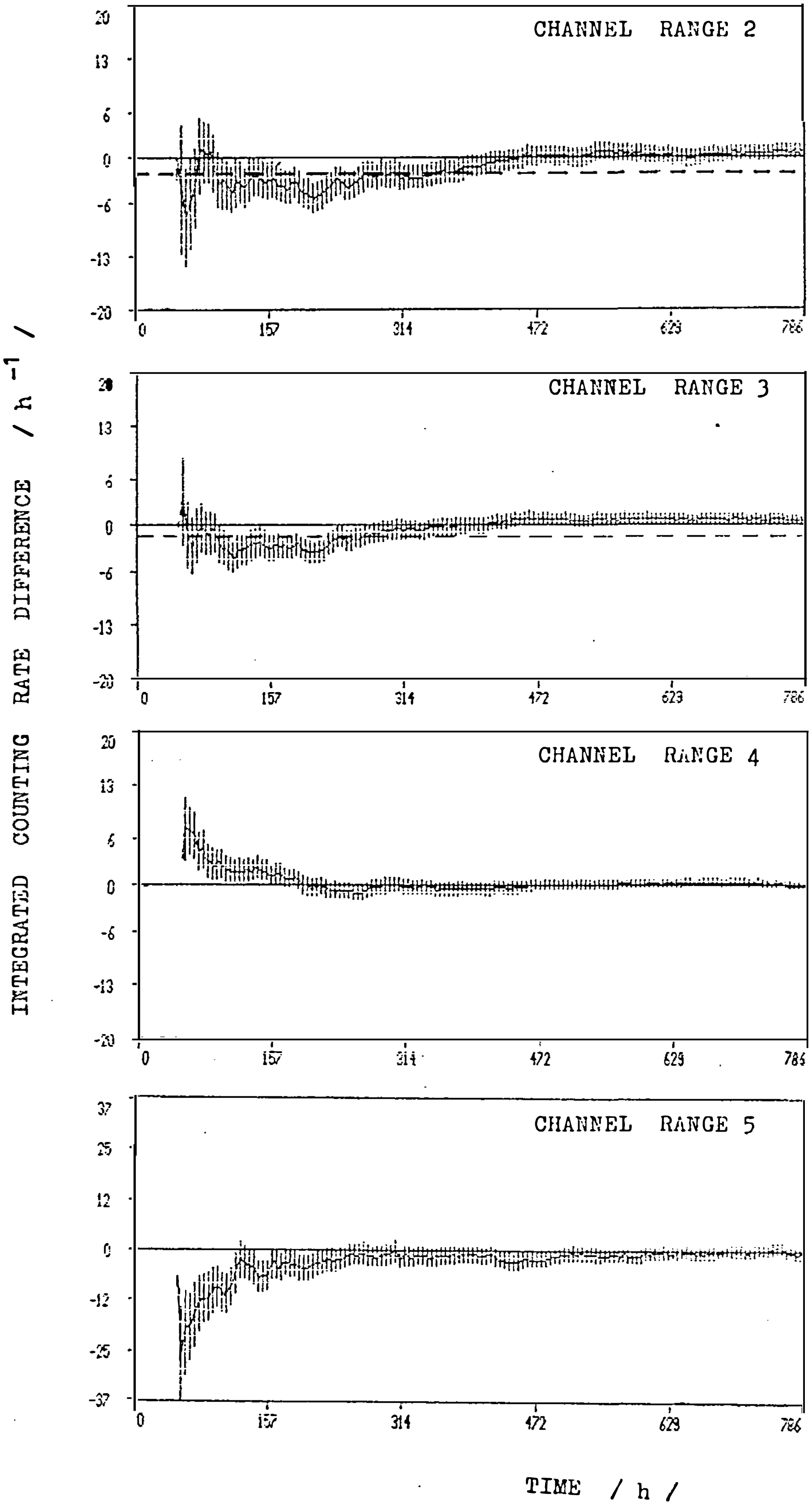

Fig. 3. Integrated counting rate difference for the channel ranges 2 3, 4 and 5 as function of running time; dd-neutrons are expected within the channel ranges 2 and 3; dashed lines — effective background level

and λ_{dd}^{Pl} — the plasma dd-fusion rate per second. For practical calculations the following parameters are used:

– $(n_D^{max}) = 6.8 \cdot 10^{22}$ [cm^{-9}] – maximum deuteron density corresponding to $PdD_{1.0}$ (which, however, is not achieved in the present experiment)
– $S_0 = 3.12 \cdot 10^{16}$ [cm^{-2} s^{-1}] – initial particle flow into the metal (corresponds to 5 mA cm^{-2})
– $V_0 = 0.2$ eV – activation energy for diffusion of D in Pd
– $T = 320$ K – temperature of electrolyte, typical for the present experiment
– $k = 8.617 \cdot 10^{-5}$ [keV K^{-1}] – *Boltzmann* constant

The loading time constant determines the gross structure of the observed experimental effects, under the assumptions mentioned above it amounts to $t_L \simeq 340$ h.

In Fig. 6 the dd-reaction rate obtained from experimental counting rate differences averaged over 60 h — time intervals for channel group 3 is shown. In this figure the shape of the model curve (1) is plotted, too. For the cell Z2 the starting time shift of 50 hours is taken into account in the model calculation. From the comparison between the absolute value of $\dot{N}^{dd} = (165 \pm 25)\,h^{-1}$ in the area of its maximum (near $t \simeq t_L$) and formula (1) we get a dd-fusion rate of

$$\lambda_{dd}^{Pl} = (1.2 \pm 0.2) \cdot 10^{-44}\, s^{-1}.$$

This is very close to the corresponding result obtained for Z1 in [6].

The integrated average reaction rate $\langle \dot{N}^{dd} \rangle$ from this model [5] is predicted as

$$\langle \dot{N}^{dd} \rangle \equiv \frac{1}{t} \int_0^t \dot{N}^{dd}\, dt = \frac{\pi}{4} (n_D^{max})^2\, d^2 l\, \frac{t_L}{t}$$
$$* \left\{ \frac{1}{3} - \exp\left(-\frac{t}{t_L}\right) \left[1 - \exp\left(-\frac{t}{t_L}\right) + \frac{1}{3} \exp\left(-\frac{2t}{t_L}\right) \right] \right\} \varkappa \lambda_{dd}^{Pl}$$
$$\sim \frac{1}{x} \left\{ \frac{1}{3} - \exp(-x) \left[1 - \exp(-x) + \frac{1}{3} \exp(-2x) \right] \right\}, \qquad (5)$$

with $x = t/t_L$.

In Fig. 7 the shape of this curve is compared to the average integrated reaction rate obtained from experimental counting rate differences for channel group 3. Again it is evident, that the gross structure of long term behaviour is reasonably described by the model prediction. From comparison between the absolute value $\langle \dot{N}^{dd} \rangle = (85 \pm 15)\,h^{-1}$ near to the end of the experimental cycle, at $t \simeq 2.16 t_L$, the dd-fusion rate can be obtained, resulting in

$$\lambda''_{dd} = (0.9 \pm 0.2) \cdot 10^{-44}\, [s^{-1}].$$

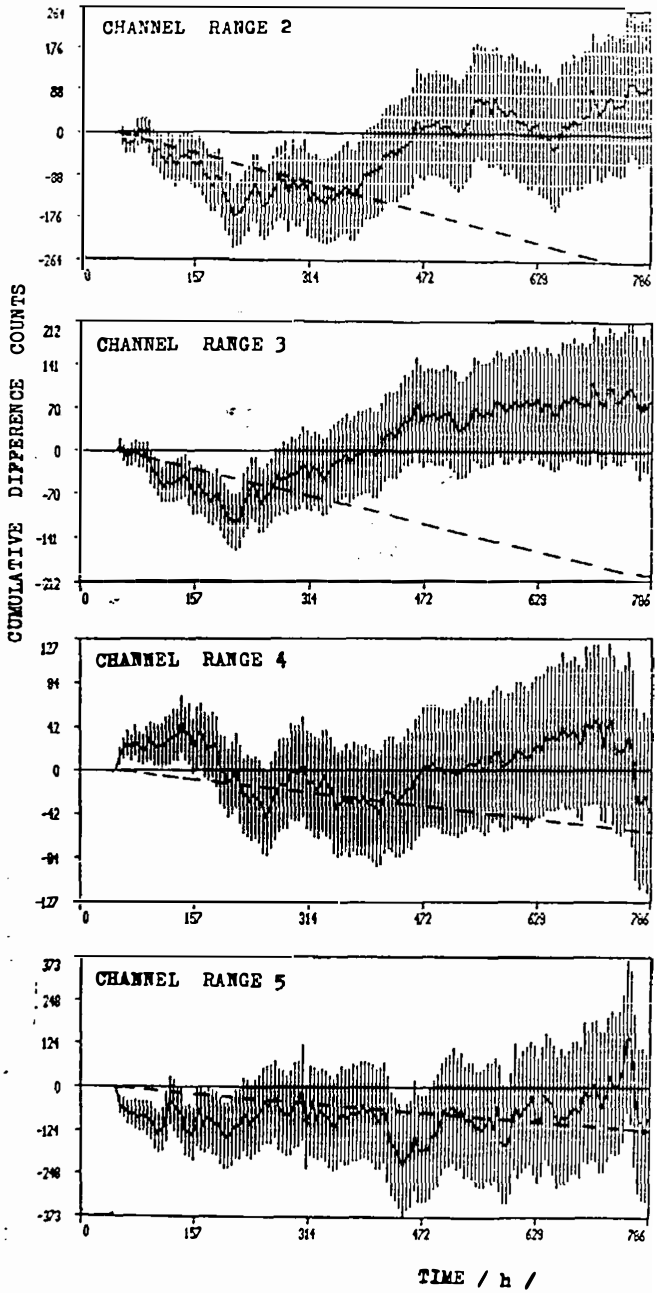

Fig. 4. Cumulative difference counts for the channel ranges 2, 3, 4 and 5 as function of running time; dashed lines — effective background level

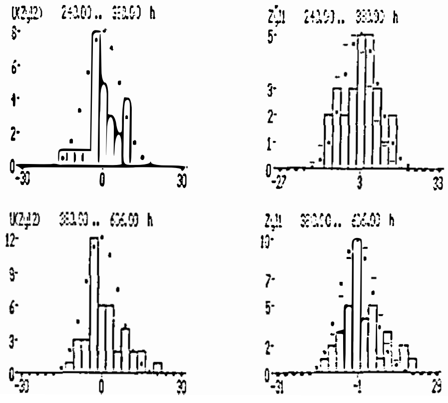

Fig. 5. Distributions of counting rate differences for cell Z2 (left side) and cell Z1 (right side) in comparison with $\varkappa^2$-minimum "background" line obtained from the measurement with Z2 for two time intervals; the measurement with Z1 in the same experimental cycle is described in more detail in paper [6]

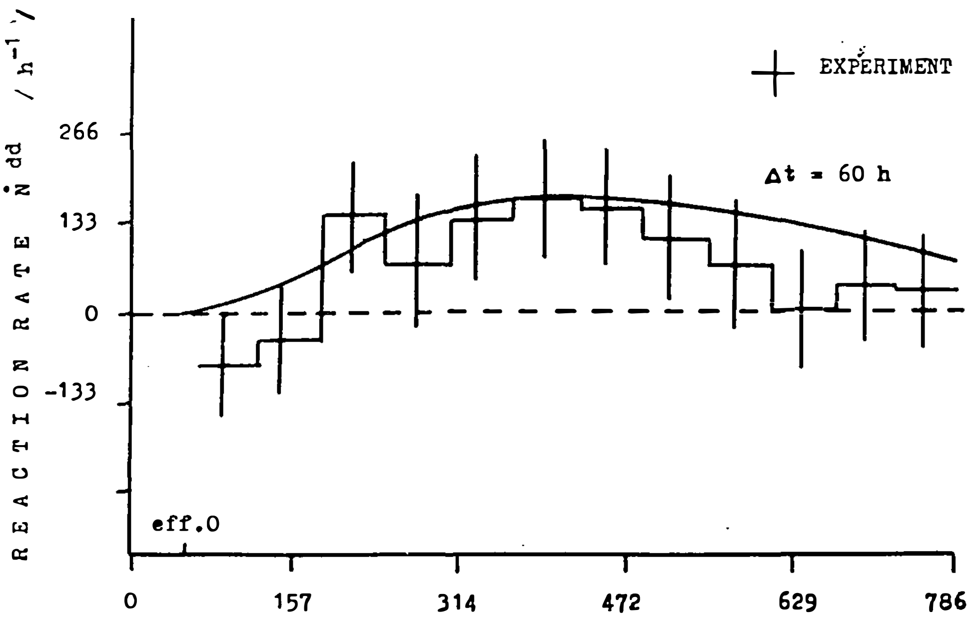

Fig. 6. Reaction rates $\dot{N}^{dd}$ for cell Z2 averaged over 60 h — intervals in the channel range 3; histogram — experiment; solid curve — calculation using eq. (1); dashed line — effective background level

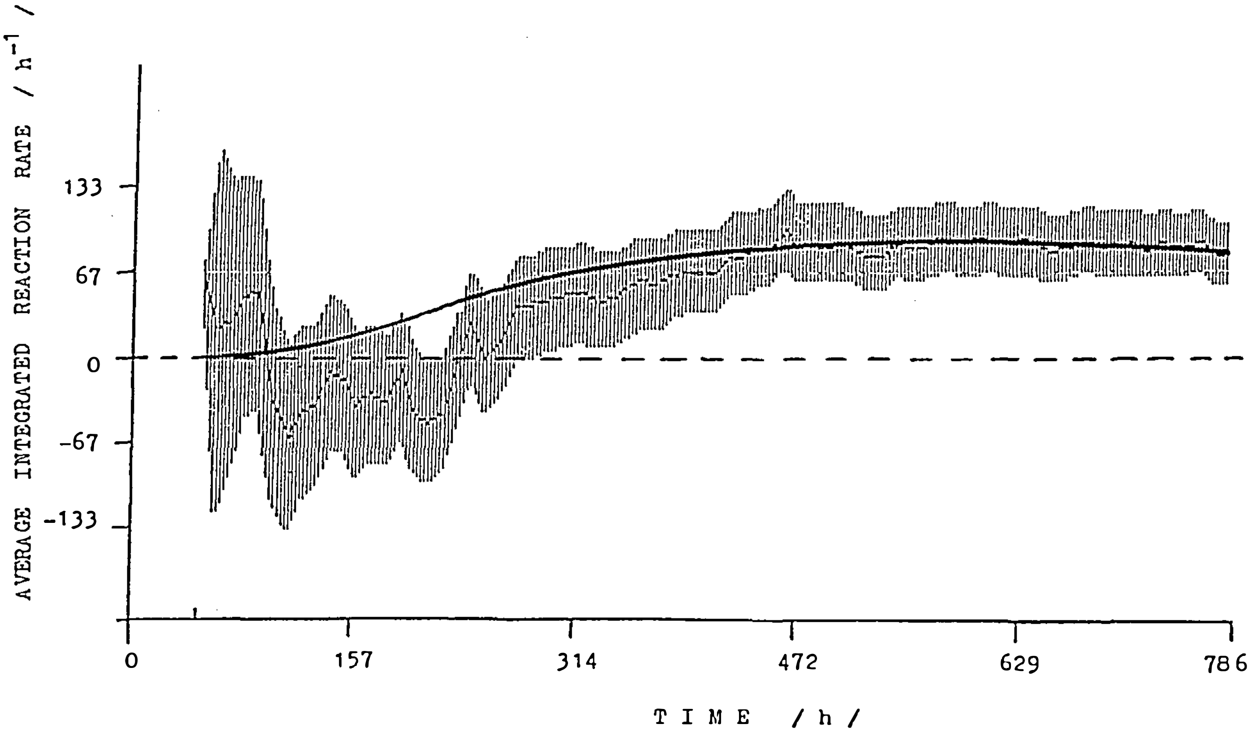

Fig. 7. Average integrated reaction rate $\langle\dot{N}^{dd}\rangle$ for cell Z2 in the channel range 3 as function of running time; the present experiment (1σ error bars) is compared with the model calculation using eq. (5) (solid curve); dashed line — effective background level

Within the estimated uncertainties both values for λ_{dd} agree between each other, although they are obtained from not identical experimental data (λ'_{dd} is determined mainly by data points up to $t = 400$ h, whereas λ''_{dd} is derived from all data points up to the end of the cycle).

The average dd-fusion rate obtained from both methods is

$$\lambda^{Pl}_{dd} = (1.05 \pm 0.15) \cdot 10^{-44}\,[s^{-1}] .$$

This is about 10% lower than the corresponding value for cylinder Z1 [6], but still in agreement with the latter within the limits of uncertainty. (For Z1 the resulting plasma fusion rate is $\lambda^{Pl}_{dd} = (1.19 \pm 0.15) \cdot 10^{-44}\,[s^{-1}]$).

5. Conclusion

The experiment with the compact palladium cylinder Z2 over more than 700 hours using a sensitive fast neutron spectrometer shows weak signals of neutrons most likely resulting from dd-reactions. However, this effect becomes observable experimentally only after more than 200 h of loading, due to the large loading time constant of such a massive electrode.

The average long-term behaviour (gross structure) of the effects observed is compatible with the predictions of the plasma-like model [5].

The resulting dd-plasma fusion rate (which per definition is equal to the product of $\langle v_d \sigma_{dd} \rangle / \mathrm{cm}^3\ \mathrm{s}^{-1}$ and unique density 1 cm^{-3} [5]) is

$$\lambda_{dd}^{Pl} = (1.05 \pm 0.15) \cdot 10^{-44}\ [\mathrm{s}^{-1}].$$

This is in the order of effects, which could be expected without assuming exotic mechanisms: In a recent paper by Scalia [8] for dd-fusion the reactivity was calculated being $1.86 \cdot 10^{-43}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$ at $T = 300$ K. The cumulative number of fusion events, which took place during the measuring cycle is in the order of $(5.3 \pm 0.6) \cdot 10^4$.

The results obtained for the cylinder Z2 are comparable with the results of investigations with a similar cylinder Z1, presented in [6].

The relative behaviour of Z1 versus Z2 shows, that active phases of both cylinders do not occur simultaneously, as it would be expected if external radiation sources would be the reason for the stimulation of dd-fusion processes. The internal non-equilibrium processes of charging the metal with deuterons, as supposed in the plasma model [5], seem to provide a more realistic scenario for the occurence of dd-fusion processes in a highly charged, compact palladium sample.

The present paper supports findings of many recent publications (see e.g. [9 ... 15] and many others) concerning the occurrence of a weak neutron production during the charging of metals with deuterons.

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