Recollecting the early exploration of nuclear reactions in condensed matter carried out in Dresden

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1. Introduction

In March 1989 Fleischmann and Pons announced the experimental observation of an unusual heat effect during the electrolytic loading of Palladium with Deuterons [1]. Besides calorimetric heat measurements, the authors also presented some measurements of nuclear radiation, which however where even less convincing, than the heat measurements. They explained this with possible hitherto unknown nuclear process, which by the authors was called "cold fusion". It is not the aim of present report to remind on all the painful history concerning announcement, exploration and discussions about this process.

Within a few weeks after this announcement, in many laboratories around the world quite different experimetal and theoretical investigations started to approve or disapprove this very unusual, but potentially extremly important phenomenon. At the TU Dresden a combined research team of scientists from the departments of nuclear physics and electrochemistry decided to concentrate the verification experiments on the search for nuclear products, mainly of fast neutrons. They had to be expected, if nuclear fusion reactions between Deuterons in the metal lattice would proceed like the well-known dd-reactions between free particles.

This is a summary report recollecting some results on the weak neutron production caused by possible deuteron-deuteron nuclear reactions in condensed matter carried out at the Technical University Dresden from April 1989 until December 1991. The aim of this report is twofold:

- It shows, that already in the starting phase of exploration some information about the evidence and mechanism of nuclear reactions in condensed matter had been obtained - just as one example for early results, which had been reported by several other laboratories too, at that time.
- The author wants to express his gratitude to all members of the former team, remembering their highly engaged work for this project - professors, senior scientists, assistants, PhD-Students and technicians.

2. Search for Neutron Emission

As mentioned, the main target was the search of fast neutrons, expected as resulting from dd-reaction (1), during the electrolytic loading of Palladium metal probes with Deuterons. The necessary techniques and experiences had been available in both institutes, therefore measurements where started very fast, already within the first half of April 1989.

Three reaction processes between free Deuterons (1-3) were well-known from accelerator experiments, one of them (3) having extremely low probability:

$$D + D \rightarrow He\text{-}3 + n + 3{,}269 \text{ MeV} \quad (1)$$
$$D + D \rightarrow T + p + 4{,}033 \text{ MeV} \quad (2)$$
$$D + D \rightarrow He\text{-}4 + \gamma + 23{,}847 \text{ MeV} \quad (\text{probability } 10^{-7}) \quad (3)$$

If considering the reactions (1) and (2), which occur with almost equal probability, then for 1Watt excess heat about 10^{12} neutrons per second had to be expected. A first rough experimental test immediately showed, that this by far was not the case: We found no fast neutrons at all. This was in accordance with calculations of tunneling probabilities through the Coulomb wall, indicating that any detectable neutron emission between free Deuterons at 0,1 ... 1 eV kinetic energy seems impossible. Further investigations could have been stopped at this point, stating a disapproval of nuclear radiations announced in [1].

But nevertheless at least two questions where still open, if for Deuterons loaded in solids due-to electron screening of Coulomb repulsion and dynamical processes in the lattice
- an upper level of neutron emission could be established in low background measurements and
- physical information could be derived about the mechanisms for such processes, if detectable.

This was the motivation for a detailed experimental program at TUD from April 1989 until December 1991 (see table 1), results of which are summarized briefly in the following.

Table 1

Experimental Cycle Series (ECS) for search of nuclear dd-ractions during electrolytic charging of metallic Palladium Electrodes at TU Dresden 1989 -1991

ECS	Date	Electrode	El. Charging /h/	No. of single 1h measurements
1	April 12 - May 03, 1989	Fr1*)	180	~ 80
2	May 22 - June 02, 1989	AH1*) Stab1**)	213 213	~ 60 ~ 60
3	June 03 - July 06, 1989	AH1*) Z1***) Z2***)	786 606 740	120 140 150
4	August 21 - September 06, 1989	Z2***) Z3***) Z4***)	368 368 368	75 75 72
5	July, 01 1990 - March 30, 1991	Z5***) Z6***) Z8***)	1250 1150 1250	651 614 64

*) - slab; **) - rod; ***) - cylinder

a) <u>First experimental series (April 12 - May 05, 1989)</u> [2,3]:

The first electrolytic cell used consists of a Palladium cathode (slab, 60x47x3 mm^3) and Platinum anode (grid, at distance 2... 5 mm), using LiOHxH_2O (0,1 M) in D_2O (99,8%) electrolyte at temperatures between 20 to 22 °C. The current density related to the geometrical surface of the cathode was 50 mA cm^{-2}. A heavily shielded neutron detector with liquid scintillation recoil proton spectrometer was used, equipped with electronic pulse-shape discrimination.

Within the recoil proton energy range from 2 to 3 MeV, as selected for registration, signals corresponding to the 2,45 MeV neutrons from the dd-reaction (1) where expected. After several hours of loading the metal probe with deuterons, the sequence of six 1h-measurements of counting effect started, followed each by equal duration background measurements. From this measurements an average effect rate of (20±5) counts per hour attributed to neutrons had been derived. Analogue measurements using H_2O instead of D_2O resulted in (-4±5) counts per hour, this means no effect. Also no effect was observed for the recoil proton energy range from 3 to 6 MeV and the same for other type of blind experiments (without current or without cell).

Taking into account detector efficiency and solid angle from the cathode to the scintillator, a weak average production rate of 0,1 s^{-1} fast neutrons, having kinetic energies below 3 MeV, in the whole volume of the Palladium sample has been determined [2,3].

For comparable masses of electrode the neutron production rate determined was by several orders of magnitude lower than the number of $4x10^4$ s^{-1} as reported by Fleischmann and Pons [1]. Therefore, it became evident, that reactions of type (1) most likely would not be responsible for any measurable heat effects. At the other side, even such small neutron production rate was much surprising, because calculations of tunneling probabilities through the Coulomb barrier at the very low kinetic energies of Deuterons did not predicted any measurable effect at all. This was considered as a first indication for possibly special properties and conditions for nuclear reactions in condensed matter. Therefore, it was decided, to continue systematic investigations of this processes in more detail.

b) <u>Method of investigation</u> [4,5]:

The special method employed for systematic studies of neutron production in condensed matter during the loading of massive metal probes with Deuterons by means of the electrolytic loading was described in detail [4,5]. It consists of the long-term observation of possible dd-reaction neutrons emanating from an electrolytic cell containing a massive palladium cathode during electrolytic charging. The neutrons are being mesured relative to the background as well as relative to other cells running simultaneously, using a fast neutron recoil proton spectrometer. Measurements relative to a cell containing light water and an others with zero electrolytic current are also included into measuring cycles. The schematic of experimental arrangement is given on fig.1. It shows the electrolytic cell, partially immersed in a water basin for cooling. Detection of slow neutrons could take place in the BF_3-counter. Fast neutrons, if emitted from the metal probe in the electrolytic cell, could be detected in the liquid scintillation recoil proton spectrometer (NE-213, 5 inch diameter, 1,5 inch high), located at a very close distance to the cell. More nuclear detectors (NaJ-, Stilbene- and plastic-scintillators) are located near to the cell, too. The whole equipment was located in a heavy iron shield, reducing the possible influence of cosmic showers and other background radiations. The scintillation detector was shielded against possible influence of magetic fields, too.

Most essential information was obtained by the recoil proton spectrometer, which was equipped with electronic pulse shape discrimination of background signals caused both by gamma rays and cosmic myons. On fig. 2 the two-dimensional distributions of detector counting rate versus light output and pulse shape amplitude are shown. Properly setting electronic discrimination biases, the signals arising from recoil protons in definite ranges of proton recoil energy can be selected. During the data aquisition proton recoil signals could be stored in up to five energy ranges, from 0,8 Mev up to 8 MeV (0,8 to 1,4 MeV, 1,4 to 2,9 MeV, 1,9 to 2,9 MeV, 2,9 to 4,1 MeV and 4,1 to 8,0 MeV), two of them including the most essential upper edge of recoil proton energy spectrum caused by 2,45 MeV neutrons corresonding to reaction (1). Only due-to using this technique it became possible to identify the extremely small effect of fast neutron emission, as reported by our group in different publications.

The method was based on simultaneous long time data aquisition of neutron signals using different cells in front of the neutron detector, as well as background measurements (empty position) and intercomparisons of counting rates between them. For the methodological studies presented in [4,5] five massive Palladium probes (weights unloaded between 89g and 172g, see table 2) were used, as volume effects had been considered to be responsible for nuclear reactions in condensed matter, at that time.

The aquisition of data for all five selected recoil proton energy ranges took place in 1h-measuring intervalls. This means, the experiments where not aimed to identify short neutron bursts, as found by some other groups. However, such intense short time events, if occurred, would have increased the counting rate measured for the corresponding 1h-intervall.

Another specific used, have been the statistical methods applied for data analyses. For instance, the distribution of counting rates, both for effect and background measurements, have been compared with Poisson distributions to get the medium rates, dispersions and variances and to check their statistical reliability (fig. 3). This procedure also was helpful to identify if any unusual behavior of measuring equipment took place during the measurement.

Further on, during the data evaluation the counts where summed up in even much broader time intervals - from 10 h up to 100 h. By this specific way of data reduction it was possible to find conditions for the optimum effect to background relation and to identify average long time trends for the process of neutron emission under investigation (fig. 4). Additionally, cummulative counting rates and counting rate differences showed the development of counting rates over the whole duration of measuring cycle, this were 370 hours for the examples presented in fig.4. The longest measuring cycle lasting about 800 hours.

Analysing the counting rates in the recoil proton energy ranges above 2,9 MeV and from long time intervals without any effect upper limits have been derived on the cosmic ray-induced neutron production and the cosmic ray myon catalyzed fusion as well, excluding this processes as possible sources of the effects observed.

Summarizing, the method used had the following advantages:

- comparatively low background counting rates in the recoil proton energy range, where signals from dd-neutrons had been expected, by using pulse-shape discrimination,
- satisfactory neutron detection efficiencies of 5 and 3 % for recoil proton energy ranges 1,4-2,9 MeV and 1,9-2,9 MeV respectively, by using a large liquid scintillator for registration,
- diminution of influence of slow drifts or time-dependent external sources of radiation because of the strictly relative procedure of measurements,
- and high sensitivity to small effects as a result of statistical methods used for data reduction, averaging and integration of counting rates.

All this specific measures finally allowed for the identification of average effects of less than 2 counts per hour with high reliability, under the background conditions in the laboratory.

c) <u>Plasma model for dd-reactions in condensed matter</u> [6, 9]:

For physical discussion of the time structure of possible dd-reaction processes a simple plasma-like model was proposed in cylindrical geometry [6] and later formlated in plane geometry [9], too. The experimental dd-neutron reaction rates $\dot{N}^{dd}$ per second in the volume ΔV of the metal electrode in the plasma model is depending on a constant plasma reaction rate λ^{Pl}_{dd} (s^{-1})

$$\dot{N}^{dd} = \tfrac{1}{2}\, n^2_D\, \Delta V <v_d \sigma_{dd}> = \tfrac{1}{2}\, n^2_D\, \Delta V\, \lambda^{Pl}_{dd} = \tfrac{1}{2}\, n^*_D\, N_D\, \lambda^{Pl}_{dd}\,, \qquad (4a)$$

where $<v_d \sigma_{dd}>$ ($cm^3\, s^{-1}$) is the plasma reactivity (product of velocity v_d and dd-reaction cross section σ_{dd} integraged over Maxwellian velocity distribution), n_D is the deuteron density, n^*_D is the density number and N_D is the total number of deuterons in ΔV. The plasma reaction rate λ^{Pl}_{dd} (4a) easily can be transferred into the reaction rate for dd-pairs Λ_{dd} (s^{-1}, per pair) frequently being used for comparisons with experiments following the equation

$$\dot{N}^{dd} = \tfrac{1}{2}\, n_D\ \Delta V\, \Lambda_{dd} \qquad (4b)$$

by recalculating $\Lambda_{dd} = n^*_D\ \lambda^{Pl}_{dd}$. The striking difference between the two presentations (4a) and (4b) is, that in (4a) reaction rate $\dot{N}^{dd}$ is proportional to n^2_D and λ^{Pl}_{dd} is constant (at constant temperature T of the electrode), whereas in (4b) reaction rate is proportional to n_D and Λ_{dd} is strongly depending on time (due-to the loading of the electrode with deuteron pairs).

In our model it was assumed, that in the metal only a part $\kappa\ [s(t) / s_o] << 1$ of all deuterons are both mobile and activated, moving through the metal and causing collisions with other deuterons located at O_h sites in the lattice,
where part of mobile particles is $\kappa = \exp(- V_o / kT)$, with the activation energy $V_o = 0{,}2$ eV for diffusion of deuterium in palladium at temperature T
and the relation $[s(t) / s_o]$ between the particle flow densities through the surface at time t and at the beginning of loading (t = 0) gives the part of activated particles.
Therefore, equation (4a) was modified for the present case of plasma rates for reactions between particles in solids into

$$\dot{N}^{dd} = \tfrac{1}{2}\, n^2_D\ \Delta V\ \kappa\ [s(t) / s_o]\ \lambda^{Pl}_{dd} = \tfrac{1}{2}\, n^*_D\ N_D\ \kappa\ [s(t) / s_o]\ \lambda^{Pl}_{dd}\ , \qquad (4c)$$

For a cylindrical electrode with diameter d and lenght l the reaction rate is

$$\dot{N}^{dd} = (n^{max}_D)^2 (\pi\, d^2\, l / 4)\ \{1 - \exp(- 4s_o\, t / d\, n^{max}_D)\}^2 \times \exp[- (4s_o / d\, n^{max}_D)\, t]\ \kappa\ \lambda^{Pl}_{dd}, \qquad (4d)$$

with the maximum particle number $n^{max}_D = 6{,}8 \times 10^{22}$ cm^{-3}, corresponding to atomic ratio D:Pd = 1. This relation (4d) was applied to experimental counting rates averaged for 1h measuring time intervals, but also for data averaged over longer time intervals (10, 20, ...100 h). By summing up counting rates from (4b), integrated reaction rates as well as accumulated reaction rates over the running time t can be determined and compared to the experiment. On fig. 5 relative shapes of three model functions comparable to experimental reaction rates are presented, pending on the variable $x = t / t_L$, with the loading time constant t_L:

(a) - average reaction rate $\dot{N}^{dd}$;
(b) - integrated average reaction rate $<\dot{N}^{dd}>$;
(c) - cumulative number of reactions $\dot{N}^{dd}_{\Sigma}$.

The model does not predict short neutron fluctuations or bursts, but the average behaviour of counting rates only, because the deuteron particle current s(t) is considered as a smooth function. However, looking at the 1h-counting rates, it is obvious, that strong fluctutions appear, most likely due-to micro-avalanches of deuterons moving into or inside the metal during the loading process. Reducing the measuring time intervals below 1h, this time-dependence of s(t) would lead to much stronger fluctuations of counting rates, finally resulting in detecting of short neutron bursts. Nevertheless, the present model was successful in describing the gross structure (the average behaviour) of reaction rates and for getting plasma fusion rates λ^{Pl}_{dd} directly derived from experimental data.

From equation (4c) one easily can conclude: The model considers nuclear reactions between inflowing mobile deuteron current s(t) with the deuterons residing on O_h sites of the target metal, the first being decreasing and the second increasing with loading time. Therefore the model predicts occurrence of a broad maximum of $\dot{N}^{dd}$ at the loading time constant t_L of a cylidrical metal electrode, this means at 63% of the full loading n^{max}_D

$$t_L = (d\ n^{max}_D / 4\, s_0), \qquad (5a)$$

where d ist the diameter of the metal electrode and s_0 [$cm^{-2} t^{-1}$] is the particle flow at t=0 of the loading process [6]. For a plane electrode (slab) the loading time constant is given by

$$t_L = (h\, n^{max}_D / 2\, s_0), \qquad (5b)$$

where h is the thickness of the slab [9]. The broad maximum of the calculated neutron emission under investigation at $t = t_L$ as predicted by the model can easily be identfied by comparing with experimental averaged counting rate differences.

d) <u>Experimental cycle series ECS2, ECS3 and ECS4</u> [7, 8, 9]:

Using the methods described, three experimental cycle series have been carried out, using five massive Palladium electrodes - AH1 (slab) and Z1...Z4 (cylinders) - and long-time electrolytic charging - 213h and up to 786 h, respectively (compare with tables 1 and 2).

The main tasks for ECS2 have been :
- Reproduction of the neutron effect observed, using different Palladium electrodes;
- studies of the production technology for Palladium electrodes;
- comparison between electrodes having extremely different masses and geometry (Stab1, AH1);
- influence of LiOD concentration on possible neutron effect;
- check if neutron signals from both cells coincide (caused by external sources of radiation?).

The main tasks for ECS3 are the following:
- Approval of positive effects for electrode AH1 as found in ESC2;
- comparison of suitability between electrodes in plane and cylindric geometry;
- comparison of neutron emission between already used slab AH1 and fresh cylinder Z1;
- Introduction into charging and observation of a second fresh cylinder Z2 with time delay of 50 h;
- study of effect rates for two almost identical electrodes Z1 and Z2;
- intercomparison of results for different electrodes as a method to distinguish between individual features and possible influences of commen external sources of radiation.

In the ECS4 three identical cylidric samples (Z2, Z3 and Z4) were observed, using different electrolytic conditions for charging. This method allowed the exclusion of electrolytic conditions, which do not lead to positive effects:
- The necessity of electrolytic current was shown with the already charged cell Z2, but running it at I = 0. This measurement showed also, that the presence of the cell in front of the detector slightly reduces the background by a small "shadow effect", which however was important for the interpretation of effects for other, active cells;
- Comparison of measurements using cell Z3 (at I = 4 A) with the measurements for Z1 and Z2 in the previous ECS3;
- Comparison of cells using D_2O based electrolyte with the measurement with cell Z4 using H_2O (at I = 4A) instead of this.

Detailed decriptions of procedures and results of measurements, data accumulation and reduction applied, comparisons with the plasma-like model and discussions have been presented separately for different electrodes in several publications [5, 7, 8, 9].

The weight of deuterons loaded in the electrodes (protons for Z4) was determined by weighing the dry probe immediately after the end of electrolytic charging during the ESC. Therefore, this number represents the average deuteron concentration over the whole volume ΔV of the electrode.

Due-to the diffusion time, which is in the order of several hundred hours, for the massive probes used there still remains a density gradient of deuterons inside of the samples by the end of ESC, whereas the density of deuterons near to the surface after very long time electrolytic loading is getting close to the maximum loading $n^{max}_D = 6{,}8 \times 10^{22}$ cm^{-3}, corresponding to an atomic ratio of x(Pd:D) = 1. Part of the experimental data, for electrodes Z3 and Z4, have been presented in [5], in connection with describing the methods used - see also figures 3 and 4.

- Selected results for AH1:

The massive slab AH1 has been used in ECS2 for 213h and - after short break of 50 h - in the ECS3 for another 786 h. In ECS2 a definite positive average counting rate difference (ACRD) within time window (100 ... 213) h of (7,0 $\pm$ 2,4) h^{-1} was observed, indicating a statistical confidence level of about 3σ. No effect was observed at the beginning, within time window (0 ... 100) h.

In ECS3 the positive effect appears within time window (205...305) h with an ACRD of (8,0$\pm$ 1,8) h^{-1} in the range of recoil proton energy (1,4...2,9) Mev and (4,8 $\pm$ 1,1)h^{-1} in the range of recoil proton energy (1,9...2,9)MeV.

The active phases of AH1 do not coincide with those of the two cells with electrodes Z1 and Z2, running in parallel to AH1. This indicates, that the effect measured does not result from external sources of radiation. Moreover, the cell with the electrode AH1 does not show positive effects outside of 1σ during the very long time interval (470...740) h, though the electrode is highly loaded and the electrolytic current of electrolyses still continues. This means, that cosmic particles, striking the detector within this time interval do not cause signals, which could be misinterpreted as caused by neutrons coming out of the cell. This also means, the dd-pairs in the fully loaded lattice do not cause a positive counting rate difference, this sets an upper limit for $\Lambda_{dd} < 1{,}1 \times 10^{-26}$ /per pair, s^{-1}/. During same time interval the electrode Z2 shows positive effects. All this experimental facts lead to high confidence in the reality of positive effects mesured for definite time intervals of the long loading process, as mentioned above.

The experimental ACRD for the electrode AH1have been compared with the predictions of the plasma-like model [6], both separately for ECS2, ECS3 and for the combined ECS2+ECS3 measurement, the later being presented on fig. 6. From χ^2 -minimum fit to the experimental data three very similar numbers for the plasma reaction rate were obtained, giving an average number of

$$\lambda^{Pl}_{dd}(AH1) = (1{,}0 \pm 0{,}15)\ 10^{-44}\ s^{-1},$$

which is in agreement with the corresponding results, obtained for Z1 and Z2.
This number has been checked by comparing to the total number of neutrons emitted from the cell during the experimental cycles ECS2+3:

- Summing up the experimental average counting rate differences in 40 h time intervals and taking into accout both the detector efficiency for the recoil proton energy range 3 and the solid angle from the cell to the scintillator, the total number of neutrons emitted within 960 h of experiment was determined, giving

$$N^{dd}_{\Sigma exp}(AH1, 960h) = (8{,}3 \pm 0{,}6)\ 10^4.$$

- Using the plasma reaction rate, the total number of neutrons emitted had been calculated by using the formulae from plasma-like model, resulting in

$$N^{dd}_{\Sigma theor}(AH1, 960h) = (12{,}5 \pm 1{,}9)\ 10^4.$$

- The experimental counting rate data for 1h measuring time interval shows strong fluctuations. The highest effect counting rate observed was $(20 \pm 5)\ h^{-1}$, this leads to (660 ± 165) neutrons, emitted from the cell. Probably, this neutrons are emitted even in a much shorter than1h time interval, most likely resulting from particle micro-avalanches inside the electrode at the time near to the loading time constant (5b) $t \approx t_L$, where the maximum of neutron activity is expected by the plasma-like model.

- Selected results for Z1 [7]:

This electrode was under simultaneous investigation over 606 h within the ECS3, together with electrodes Z2 and AH1. It showed weak, but definite signals of neutrons that most likely result from dd-reaction (1). The effect became observable after >200 h of loading only, because of the large loading time constant of such massive electrode. After this, strongly fluctuating positive effects occured during several hundred hours with slowly decreasing intensity. Within time interval (285...315) h the average counting rate difference to the background for the most important recoil proton energy range (1,9...2,9)MeV was $(9,5 \pm 2,1)\ h^{-1}$, this corresponds to 4σ statistical confidence. The highest single counting rate difference amounts to $(30 \pm 10)\ h^{-1}$.

The average long-term behavior of the effects observed was compared with the predictions of the plasma-like model [6] as shown on fig.7 for the experimental averaged CRD within 60 h intervals, using the calculated loading time constant $t_L = 350h$. Adjusting the amplitude of theoretical curve to the experimental distribution the plasma fusion rate was determined. Additionally, the plasma fusion rate was determined by comparing the model curve to the experimental average integrated counting rate $\langle N_{dd} \rangle$ resulting in an very close number. The average number from both methods, finally adopted, was

$$\lambda^{Pl}_{dd}(Z1) = (1,19 \pm 0,15)\ 10^{-44}\ s^{-1}.$$

The fluctuating character of the effects indicate, however, that the particle flow inside the metal was not a continuous, smooth as assumed by the model, but rather behaved as resulting from statistically distributed micro-avalanches, transporting the deuterons into the depth of the metal.

The cumulative number of neutrons from dd-reaction events that occurred during the measuring cycle was in the order of $(6,26 \pm 1,1)\ 10^4$.

- Selected results for Z2 [8]:

The cylindrical sample Z2 during the ECS3 was loaded with deuterons electrolytically for 736 h in measurements parallel to measurements with the two other cells containing electrodes Z1, AH1 and to background measurements (empty position in front of the spectrometer). The measurements showed weak, but definite signals of neutrons most likely resulting from dd-reactions. However, the effect became observable experimentalls after more than 200 h of loading only, due-to the large loading time constant $t_L \approx 340h$ of this massive electrode.

The reaction rates averaged over 60h-intervals and average integrated reaction rates have been compared with the plasma-like model, as shown on figs. 8 and 9. The average plasma fusion reaction rate obtained from both methods is

$$\lambda^{Pl}_{dd}(Z2) = (1,05 \pm 0,15)\ 10^{-44}\ s^{-1},$$

this is about 10% lower than the corresponding number for Z1, but still agrees within the limits of uncertainty. It is also in accordance with the result for AH1, though the geometry of Palladium electrodes for both cases is completely different.

The cumulative number of reaction events which took place during the measuring cycle is in the order of $(5,3 \pm 0,6)\ 10^4$. This is in agreement within the uncertainty limits with the result for Z1, having almost same mass and dimensions as Z2. The corresponding result for AH1 is about two times higher, probably due-to the almost two times higher mass of that electrode.

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 sources of radiation would have stimulated dd-reactions in both samples. The internal non-equilibrium processes of charging the metal with deuterons seem to provide more realistic scenario for the occurrence of dd-reaction processes, as proposed by the plasma-like model [6].

e) <u>selected results from ECS5 (electrodes Z5, Z6, Z8)</u> [13]:

The experiments carried out in ECS5 were designed for the following purposes:
- Investigation of time dependence of counting rates using essentially improved sensitivity of the experimental arrangement;
- study of the infusion process of the Deuterons including its non-equilibrium characteristics;
- exploration of the influences of metal surface characteristics on both infusion and electrolyses;
- determination of Tritium enrichment in the electrolytes during the electrolyses pocess;
- determination and unfolding of recoil proton effect spectra to get information about the neutron energy emanated from the electrodes.

For characteristics of the electrodes used - see tables 1 and 2.

Table 2

Data of the massive Palladium samples used in ECS1-5 *(foto: Fig. 12)*

electrode	*dimension (mm)*	*weight Pd (g)*	*current (A)*	*electrolyte*	*weight D*$^{*)}$ *(mg)*	*av. ratio D/Pd*	*description references*
Fr1	60x47x3	102,0	6 / 3	0,1 M LiOH D_2O (99,8%)	n.d.	-	[2, 3]
Stab1	D5,0 L25,0	26,3	2,3	3 M LiOD D_2O (99,8%)	n.d.	-	
AH1	49,8x40,3x8,4	171,71	8	"	2286	0,698	[9, 5]
Z1	D23,1 L19,3	93,54	4	"	1419	0,80	[7]
Z2	D22,6 L20,2	92,69	4	"	1424	0,81	[8, 9, 5]
Z3	D22,6 L18,6	89,02	4	"	1420	0,826	[5]
Z4	D22,6 L18,7	89,9	4	3 M LiOH H_2O	720 (H)	-	[5]
Z5	D22,2 L18,3$^{**)}$ D_i 2,0 L_i15,0	85,78	4	1 M LiOD D_2O (90%)	1419	0,94	[13]
Z6	D22,2 L18,1	86,33	4	"	1118	0,75	"
Z8	D34,8 L 45,5	518,21	8	"	7387	0,81	"

$^{*)}$ maximum weight of D at the end of el. loading process;

$^{**)}$ hollow cylinder, with an inner diameter of 2 mm to extract gas out of the sample and for studies of non-equilibrium process in the sample

The electrode Z6 was used for comparisons with results from previous measurements in ECS3&4. The probe Z5 was a hollow cylinder, aimed on the study of deuterium diffusion through the metal and on establishing of a stationery state of permanent particle flow through the cylinder. The cylinder Z8 was much bigger than previous samples and aimed for the tests of both surface and volume effects of the neutron production investigated.

The experimental arrangement for ECS5 as shown on figs. 14 and 15 had been extended and improved essentially comparing to the arrangement used in previous ECS2-4:

- Three neutron detectors (marked with R, E and T), each with NE-213 scintillators, are used for the recoil proton spectrometry. This measure improves the counting statistics and reliability of results of experiment (fig. 14).
- The cell holder comprises a cylindrical turning box (filled with water), which allows to shift each of three cells and the empty position (background) to the front of each of the three detectors. By means of this installation the processes in each cell can be observed continously, but using different detectors. This allows for a consequently relative type of measurements, comparing the counting rates between each pair of cells and/or of each cell with background (empty position).
- The whole arrangement of detectors, cells and turning cell holder is covered by a massive background shield, mainly consisting of polyethylene blocks (fig. 15). Using tests with a Cf -252 neutron source the reduction of background level in the most essential recoil proton area around 2-3 MeV down to 20-30% of previous level was reached.

The analyses of experimental data for ECS5 comprises of following procedures:

- Statistical analyses of measured counting rates corresponding to definite energy intervals and investigation of time dependences for average integrated and cumulated counting rates (similar to the procedures pesented above for ECS3 and 4). Similar results are obtained as for previous ECS concerning the time-dependence of neutron effects, as described by the plasma-like model.
- Determination of averaged recoil proton spectra for definite time intervals of measurements and analyses of this data concerning their shape, position of upper edges, average neutron rates and other criteria (fig. 16). Finally by unfolding methods the shape of neutron spectra have been determined, showing broad maxima at neutron energy of (2,6 $\pm$ 0,2) MeV (fig. 17 and table 3).

Table 3

Data of neutron spectra reslting of unfolding of the recoil proton energy spectra for different electrodes Z5, Z6 and Z8 used in ECS5 [13]

Electrode	**medium neutron energy /MeV/**	**half width ΔE /MeV/**	**neutron rate in the spectrum $\dot{N}_s$ /h^{-1}/**	**neutron rate produced in cell $\dot{N}^{dd}$ /s^{-1}/**
Z5	2,6	0,2	35 $\pm$ 6	0,1 $\pm$ 0,02
Z6	2,5	0,2	58 $\pm$ 9	0,16 $\pm$ 0,03
Z8	2,6	0,2	46 $\pm$ 10	0.13 $\pm$ 0,03
Average	2,6	0,2	49 $\pm$ 11	0.14 $\pm$ 0,03

The results of ECS5 analyses strongly support the conclusions from analyses of previous ECS concerning the evidence of dd-neutron emission with an average rate of $\dot{N}^{dd}$ = (0,14 $\pm$ 0,03) s^{-1} resulting from nuclear dd-reactions in Palladium during the non-equilibrium loading processes of Deuterium. Taking into acount, that cells have been charging during ECS5 for more than 1000 h, that result means, that more than 5x10^5 dd-neutrons have been produced in each cell!

f) <u>neutrons from loading of Ti from the gas phase</u> [10]:

In a deuterium gas loading experiment 58g of Titanium turnings in a stainless steel container where used. The container could be alternatively heated, evacuated or filled with Deuterium gas. The neutron emission was studied during the absorption process of Deuterium gas by the Titanium turnings in two experiments. In both experimental runs definite signals of weak neutron production where observed. The fusion rate per dd-pair derived from average and maximum effects has been equal to $\Lambda_{dd} = 6{,}6\ 10^{-25}\ s^{-1}$ and $\Lambda_{dd} = 2{,}67\ 10^{-24}\ s^{-1}$, respectively. However, this fusion rate numbers are for non-equilibrium conditions of the loading process, not for the stationary fully loaded Ti sample, because in the later case no neutron counts are observed at all.

The recoil proton spectrum is in accordance with the assumption, that dd-neutrons have been detected in this experiment (fig. 10). Some weak indication for neutrons at higher energies is also present, but with confidence level of 1σ only.

The correlation between effect counting rates and both gas pressure p and change of pressure dp was investigated. It was found, that the strongest correlation was observed for the product p x dp, that means to the product of already absorbed deuterons and the additional flow of particles through the surfaces into the metal. This is in general agreement to the findings of the plasma-like model. The direct application of this model was impossible however, due-to the complicated geometry of the Titanium turnings.

g) <u>neutrons emanated during degassing of Pd probes</u> [11, 12]:

A very big Pd cylinder Z8 (D34,7 mm, L45,4 mm, 518,207 g) was loaded with 8 A in 1M LiOD solution in D_2O (90%) for 1254 h with 7,387g of Deuterium, which corresponds to an average atomic ratio Hydrogen:Palladiumd of x=0,8 $\pm$ 0,01. In a special experimental device this sample was heated to push out the deuterons from the metal and observed with two neutron spectrometers in 18 short measuring runs over 264 min totally. After each run, the Pd sample position was turned from one to the other detector. Follwing this procedure, each detector had measured alternating effect as well as background counts.

The recoil proton spectrometers anabled a crude neutron spectroscopy. Both detectors showed a weak but definite effect-background difference for recoil proton energy range 1,9 ... 3,3 MeV, whereas in the range 3,3...5,2 MeV there is almost no difference. The recoil proton spectrum for 0,5 MeV bins obtained, is in accordance with an experimental spectrum of dd-neutrons, measured at a neutron generator (fig. 11).

The neutron emission observed is pending on nonequilibrium conditions: No neutrons are observed from fully loaded sample before the start of expelling the gas by heating. During degassing, the counting rate difference is decreasing rapidly. The reaction rate per dd-pair determined for the highest effect rate is $\Lambda_{dd} = (3 \pm 1)\ 10^{-25}$ per dd-pair and per second, which is in the same order as found for previous experiments for Ti turnings. This fusion rate characterizes the non-equilirium conditions during degassing of Pd sample, not the situation for the fully loaded sample before degassing was started by heating.

3. Search for Tritium production [13]

The ECS5 also aimed on the determination of Tritium potentially being produced by dd-reaction (2) during the electrolytic loading of Palladium samples with Deuterons. There are two methods, which could be used for this purpose: Either analyses of gas expelled from the cathodes after the end of electrolytic loading, or analyses of the electrolyte, where Tritium is accumulated also by permanent ion exchange with the Hydrogen (both Deuterium and Tritium) content in the cathodes during the electrolytic process. The second method was used here, as the very long electrolyses in ECS5 of more than 1000 h was sufficient for the ion exchange process between gases included in the cathode and the electrolyte. The Deuterium used for the composition of electrolyte already has some Tritium content, pending on the production method of the Deuterium. This T-activity is considered as the background/starting number of T-content.

Table 4

Results of measurement of the enrichment factor α of Tritium activity in electrolytes used for ECS5 (derived from table 4.14 [13])

cathode	**probe Nr.**	**electrolyses time [h]**	**T activity [dpm/g]**	**enrichment factor α**
fresh electrolyte	1	0	199 ± 8	≈1
"	2	0	192 ± 8	≈1
"	6	0	208 ± 8	≈1
"	7	0	201 ± 8	≈1
"	12	0	185 ± 8	≈1
Z5	4	1006	327 ± 10	1,68 ± 0.03
"	9	1099	345 ± 10	1,77 ± 0,06
"	14	1194	367 ±16	1,88 ± 0,09
"	17	1269	390 ±13	1,75 ± 0,07
"	20	1360	408 ± 9	1,87 ± 0,10
Z6	3	1006	330 ± 6	1,69 ± 0,05
"	8	1099	265 ± 13	1.87 ± 0,08
"	11	1170	424 ± 8	1,94 ± 0,04
Z8	5	1006	328 ± 8	1,68 ± 0,04
"	10	1099	328 ± 11	1,68 ± 0,06
"	15	1194	346 ± 11	1,77 ± 0,06
"	18	1269	399 ± 8	1,82 ± 0,01

During electrolyses, part of the D_2O is splitted into D_2 and O_2 gases, permanently. This losses of electrolyte have to be replaced during the long electrolytic time, either by a permanent low inflow of fresh D_2O or by adding equal portions of fresh D_2O periodically. Second method was used here. Theoretical consideration of accumulation process shows, that the Tritium content in the electrolyte for both cases of refilling reaches a saturation level, which is higher than the Tritium concentration in fresh electrolyte by an enrichment factor α, pending on the production rate of Tritium in the cathode by the reaction (2).

For analyses of Tritium activity small probes of both fresh and used electrolyte (after more than 1000 h of electrolyses) were collected and analysed in a liqiud scintillator spectrometer for low level counting at the Bergakademie Freiberg (today: Technical University Bergakademie Feiberg), where this method was well-established and available for geo-radiological monitoring. Such analyses were carried out several times both for fresh electrolyte (background) and for each cathode Z5, Z6 and Z8, as used in ECS5. From Tritium counting rates of the spectrometer the specific Tritium activity has been calculated. Results of the measurements and of theoretical modeling of the enrichment process are given on fig. 18 and in table 4. After about 800 h of electrolyses the enrichment factor α (t-activity in the active electrolyte divided by its starting/background level) reaches almost the same numbers for all cells Z5, Z6 and Z8. From the experimental investigation with all three samples a definite indication about the production of Tritium during the electrolyses process was obtained, most likely resulting fron the dd-reaction (2).

By a very conservative consideration of tritium activity, calculated from the measured counts of Tritium decay, a total Tritium production rate of $\dot{N}_T = (1{,}24 - 0{,}54)\ 10^4\ s^{-1}$ in the whole volume of cathodes was determined. Comparing to the dd-neutron production rate of $\dot{N}_n = 0{,}14\ s^{-1}$ found in ECS5 too, this means, that the relation between Tritium and Neutron channel of dd-reaction was in the order of $\dot{N}_T/\dot{N}_n = (8{,}9 - 3{,}8) \times 10^4$. This result was in qualitative agreement with results of Tritium analyses in other electrolytic experiments at that time, which showed an enhanced relation of Tritium:Neutron production even in the range 10^6 - 10^9 (*Authors addition from todays position: A less conservative but more realistic consideration of Tritium determination for ECS5 experiments would lead to a $\dot{N}_T{:}\dot{N}_n$ relation in the range 10^5 ... 10^6, that is close to the lower level of results observed by other groups*).

This results for the splitting between tritium and neutron channels of dd-reaction showed once more, that nuclear reactions in condensed matter essentially distinguish from reactions between free particles.

4. Reviews with an attemp for explanation [14, 15, 16]

Detailed reviews of both experimental and theoretical work on nuclear reactions in condensed matter, as published by the end of 1989, as presented in [14, 15]. In the conclusions of this papers the importance is stressed for future investigations of time-dependent description of Coulomb microfields inside of the solids. The description of the tunneling process through dynamical Coulomb potentials was mentioned as still being an open problem. Non-equlibrium processes could lead to an increase of fluctuations, which are enhancing tunneling probabilities, too.

In an other review the theoretical limits of nuclear fusion reactions in condensed matter where considered [16], by August 1990. Hypothetical enhancement of dd-reactions by both static and dynamic effects and fluctuations have been considered in theoretical publications at that time. Basing on this papers presented by other authors, the review came to the following conclusions:

- The free D_2 molecular fusion rate is more precisely estimated in the order of $\Lambda_{dd} \approx 10^{-64}\ s^{-1}$;
- strong enhancement takes place due-to static screening by electrons in the valence band with a factor $f_{sc} = 10^{10}$ ... 10^{15};
- dynamical effects and fluctuations may further increase fusion reaction rates by a factor of $f_{fl}^{Pot} \approx 10^6$ ... 10^8, or even higher;
- this results in realistic estimates of dd-fusion reaction rates in the metal lattice within the range $\Lambda_{dd} \approx (10^{-46}$ ... $10^{-29})\ s^{-1}$, under special conditions further enhancement is not impossible.

The reaction rates observed by measurements of neutron emission at TUD during the infusion of deuterons into metal probes (as well as from degassing), by many orders of magnitude where smaller than numbers suggested in the original paper by Fleischmann and Pons [1], but they where still much higher than numbers expected for free D_2 molecules, without assuming any special physical mechanisms enhancing this rate by the influence of dynamical solid state or screening processes.

In the summary of the review [15] it was written: *"No doubt, the field of nuclear fusion reactions in condensed matter still is in the phase of its beginning. However, already now it raised interesting physical problems and it gave insights into processes, which so far have almost not been considered"*. Further detailed studies of nuclear reaction mechanism in condensed matter where recommended in this review.

5. Looking back from 2021

Just 30 years passed, since the investigations of neutron emission from dd-reactions in condensed matter by the TUD team had to be terminated. Since that long time, a tremendous amount of experimental information on this emerging field of nuclear processes in condensed matter was collected worldwide by many research groups, as it is documented for instance in the library of the CANR organisation [17], by publications in the Journal of Condensed Matter Nuclear Science [18] detailed reviews [23-27] and in a huge amount of original publications. The generation of unusual heat was reproduced many times by independent researchers, also Helium, Tritium and Protons from dd-reactions where measured.

Moreover, the LENR area extended very fast: It was shown, that proton induced LENR processes are occuring, too. Even transmutations and isotope changes of heavy nuclei have been observed many times. Experimental methods for activation of such processes diversified: Besides the traditional electrolytic loading, also absorption of hot Hydrogen gases in solids, electric discharges in gases or vacuum and acoustic methods (cavitation) became used successfully. Nevertheless, because of missing accepted theoretical description, the LENR research was not fully recognized by official science community, for nearly three decades.

With high interest the world physical community noticed two recent publications in the APS journal Physical Review C by a NASA research team in spring 2020, which describe both experimental and theoretical investigations of nuclear reactions in metal lattices [19, 20]. In the abstract of paper [20] it is stated: *"Nuclear fusion reactions of D-D are examined in an environment comprised of high density cold fuel embedded in metal lattices in which a small fuel portion is activated by hot neutrons. Such an environment provides for enhanced screening of the Coulomb barrier due to conduction and shell electrons of the metal lattice ... In general, the effect of screening becomes important at low kinetic energy of the projectile. ... We demonstrate that for* $E \leq U_e$*, a direct calculation of Gamow factor for screened Coulomb potential is required to avoid unreasonably high values of the enhancement factor f(E) by the analytical — and more so by the asymptotic — formulas."*

Another promising approach to the theoretical description of nuclear reactions in condensed matter has been presented by Kalman and Keszthelyi [21, 22] (and many other publications of the same authors). They consider the Coulomb interaction between Deuterons (or Protons) moving in the metallic lattice both with the free electrons in the conduction band and with electrons bound to the atomic shells in the lattice as a 2-nd order perturbation to the nuclear interaction with other heavy particles (Deuterons, Protons a.o.) included in the lattice. As a consequence of this perturbations the following 3-particle process between Deuterons becomes possible

$$^{A}_{Z}L + d + d \rightarrow {}^{A}_{Z}L` + {}^{3}He + n + 3{,}269\ \text{MeV}, \qquad (6)$$

where $^{A}_{Z}L$, $^{A}_{Z}L`$ are the lattice atoms, which catalyse the nuclear reaction by Coulomb interaction, but which do not participate in the nuclear reaction directly. The authors state: This type of three-body interaction explains the two main empirical observations of LENR, their occurrence and the absence of energetic Gamma-radiation. It was found that electron catalyzed process producing He-4, like in reaction (3) for free particles, by far has the largest rate among possible electron catalysed dd-processes. This theoretical approach could also explain the magnitude of fusion reaction rates λ^{Pl}_{dd} as found in our experiments [3-5, 7-9].

In the theoretical review by Kalman et al. [22] it was stated, that *"it is possible to consider protons and Deuterons on and within metals and alloys as type of plasma due to their high mobility. ... In the interior of metals where the fields usually are shielded, diffusion can still produce relatively rapid motion of hydrogen isotopes. ... That mobility has clear foundation. Hydrogen isotopes in lattice do not have bound electrons ... this gives them sizes on the scale of femtometers, the normal nuclear size scale. The small size is the root of their very high diffusion coefficient.... Protons and Deuterons in materials that exhibit LENR are much like positive ions in ordinary plasmas with a significant exception. Their motions are not random but constrained by directions available in the plasma. However, they are still very mobile"* (citation from [22], page 16).

This actual statement is in agreement with the assumptions made in our plasma-like model for the description of neutron counting rates during the infusion of Deuterium into Palladium [6].

Recollecting the exploration of fast neutron emission during electrolytic loading of Palladium with Deuterons, carried out at TUD about 30 years ago, the following conclusions can be drawn from a topical position:

- Using massive metal probes and long time charging with Deuterons a very weak neutron emission became observable, most likely resulting from nuclear dd-rections in solid environment due-to the electron screening of the Coulomb repulsion.
- This process was studied systematically, showing its dependence both from density of Deutrons n_D and from density derivation dn_D/dt, which in the frame of a plasma-like model is interpreted as an interaction between the active part of mobile Deuterons, flowing into and inside (but also out of) the metal, and the Deuterons residing in the metal lattice.
- It was shown by the voluminous number of investigations worldwide, that the neutron emitting channel of dd-interactions in condensed matter is smaller by many orders of magnitude comparing to the production of He-4 or Tritium. This behaviour was found in the present investigatons of Tritium content in the electrolyte, too. Nevertheless, neutron detecion might be important for understanding of the reaction mechanisms, because it delivers information about the time-dependence of the processes under investigation even for massive reactors or metallic samples, that cannot be obtained from analyses of components in gases, radioactivity in liquids or traces in track detectors.
- No doubt, the generation of nuclear dd-reactions in condensed matter by using the flow of active Deuterons in a metal lattice already partially loaded with Deuterons is one way to detect such processes and to study the influence of electron screening on the Coulomb interaction in the metal environment. However, using this method, the time window in which measurable dd-processes can be expected is limited to the duration of 2-3 loading time constants t_L. (However, organizing in the experiment a permanent flow of deuterons through a loaded metal layer, this disadvantage of limited time window for reactions could be avoided).

  An other way avoiding this limitation in time is the activation of dd-processes in fully loaded solid by an exteral source of radiation, as recently presented by the NASA group [19,20].

  Recently, even transmutation of elements by long time pressure cycling of Deuterium through Palladium Silver metals was observed [28].

  Generally speaking, for this type of investigations of nuclear reaction processes in condensed matter, where necessary initial conditions of solid state lattice and the distribution of fuel in it (Protons, Deuterons, Tritons) are reasonably known, the physical processes are easier to understand. It`s helpful in particular, if all competing reaction channels could be investigated simultaneously. This path of investigations is on a good way to the complete understanding of the specifics of nuclear reactions in condened matter, for particular conditions of reasonably known electron environment in metallic solid for the reacting nuclear particles.

- The present field of LENR is much broader, however. It includes processes, which occure due-to local properties of the solid (cracks, lattice defects a.o.), formation of different condensates of the fuel particles on the surface or inside of the solids, nanostructures of metals or alloys, coherent excitations of either fuel particles or lattice atoms, special electromagnetic or phonon excitations (electric discharges, formation of plasmon polarons, resonances, collaps of cavitation bubbles near to metal surfaces), special chemical processes on the surface, catalysing the condensation processes between fuel particles and others. Moreover, many experimental investigations are focused on the main result of LENR only, this is the generation of unusual heat (because this is of relevance for potential application), missing therefore a complete picture about all processes in their experiments. No doubt, also this explorations of LENR will take advantage of results from experiments carried out in well-understood solid state environment, as mentioned above, and also of the theoretical results about general influence of both conduction and shell electrons in metal lattices on nuclear processes in condensed matter (as proposed f.i. in [20, 21, 22]).

The complete theoretical understanding and descpription of each approach to LENR in quite different electron environment deserves separate specitic theoretical considerations, first of all based on well-established physical principles, instead of unapproved hypotheses.

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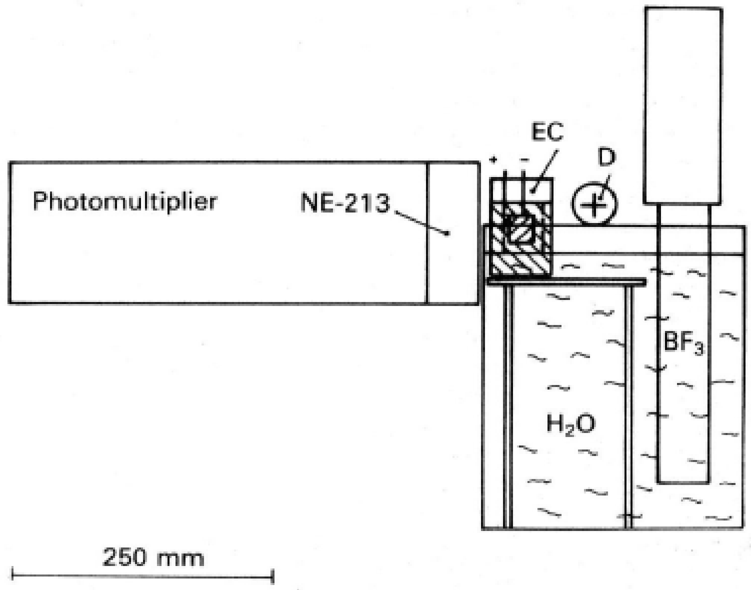


Fig. 1 Schematic of experimental arrangement: EC – electrolysis Cell; H_2O – water basin; D – position of various other detectors; the heavy iron shield is not shown

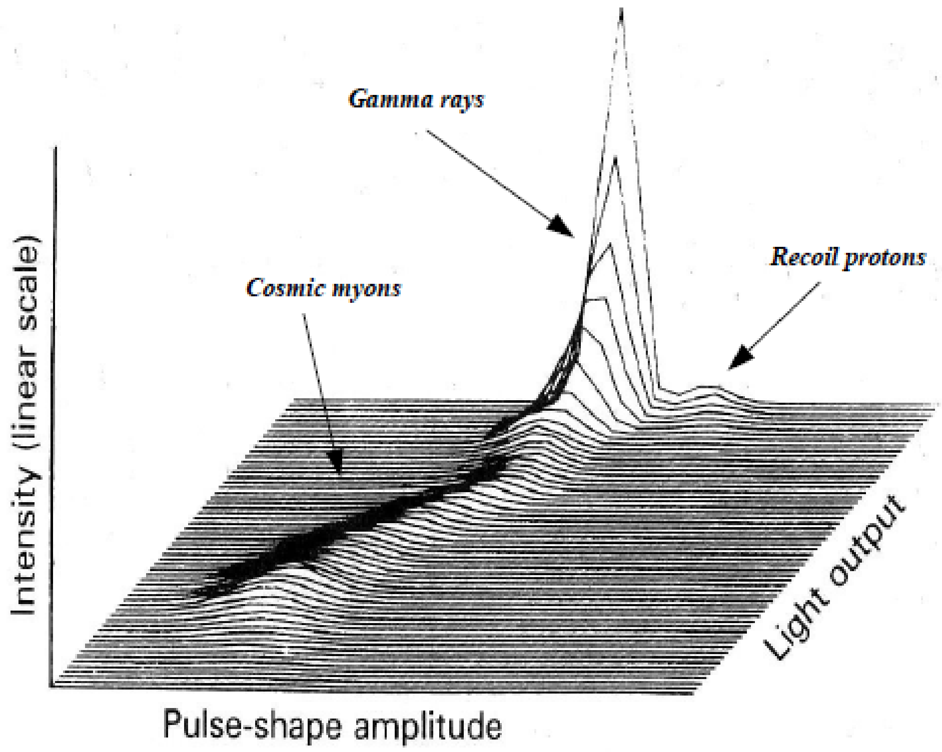


Fig. 2: Three dimensional background spectrum versus light output and pulse-shape signal amplitude for the recoil proton spectrometer used. Very few fast neutrons are contained in the background spectrum only.

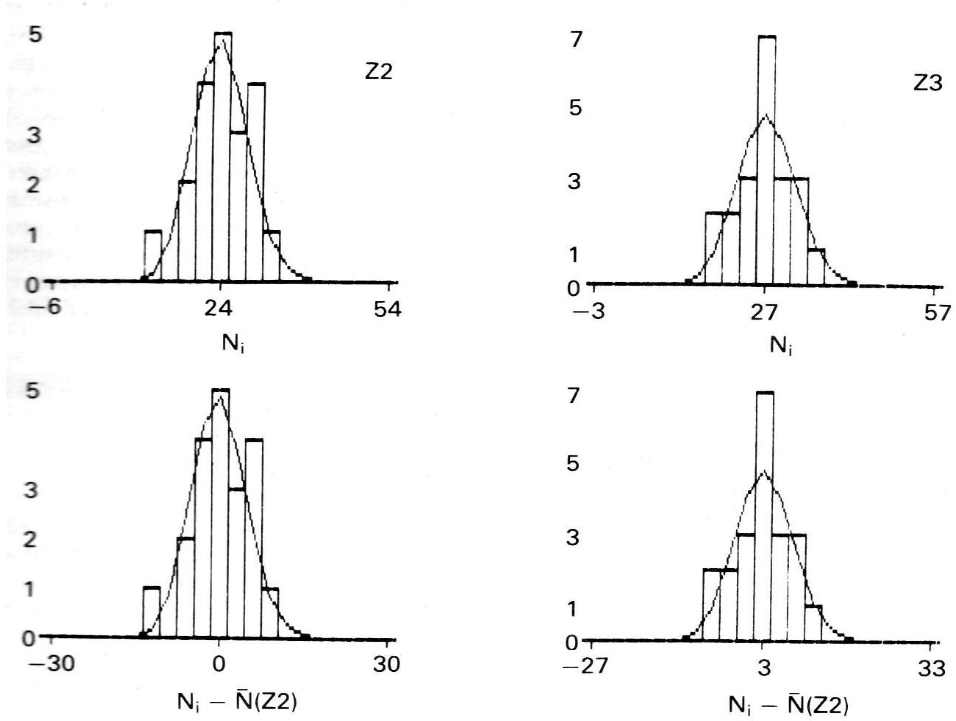


Fig. 3: Distributions of 1h counting rates N_i in recoil energy group 1,9...2,9 MeV for palladium probe Z2 (without electrolytic current) and Z3 (with Deuterium loading) within the time interval from 100h to 200h of electrolysis (upper part) and relative to an averaged background line obtained from the Z2-masurement (bottom). The Z3 measurement shows an average effect count of 3 h^{-1} over 100 hours comparing to Z2 probe. Smooth lines represent Poisson distributions.

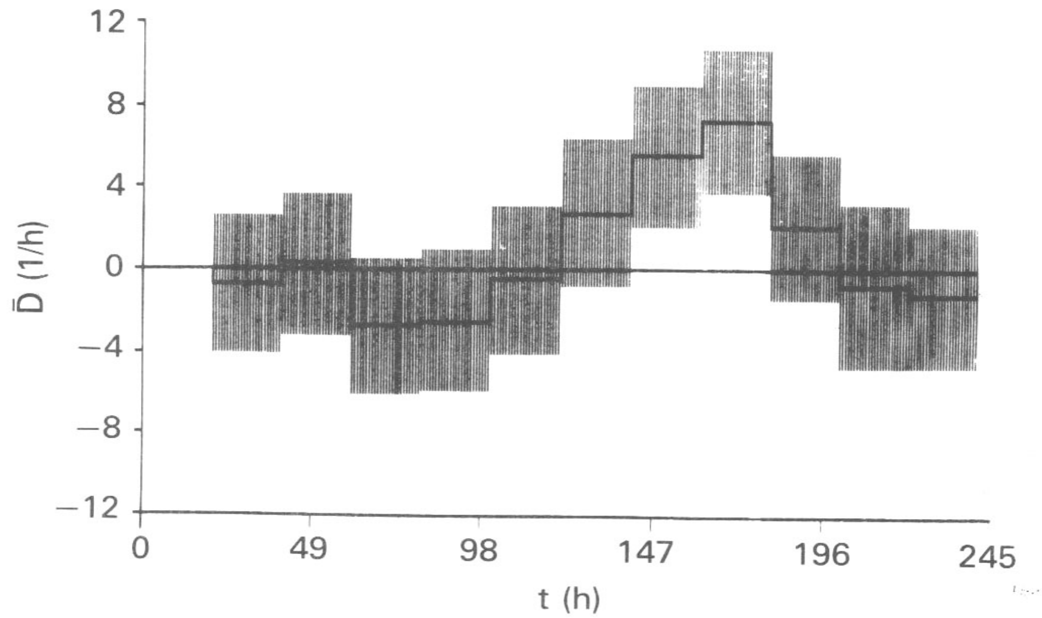


Fig.4: Average counting rate differences for Z3 – Z2 using same data as on fig.3. Counting rates for Z2 and Z3 are summed up in 20 h time intervals. The difference clearely shows an effect counting rate between 100 h and 200 h of measuring time (Z3 electrolysis time) for proton recoil energy range 1,9 … 2,9 MeV, most likely produced by dd-neutrons following reaction (1)

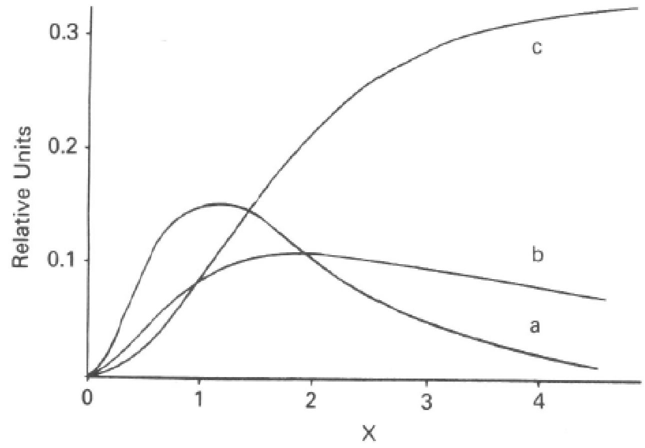


Fig. 5: Relative shape of functions obtained by the plasma-like model comparable to experiments; a) – average reaction rate; b) – integrated average reaction rate; c) – cumulative number of reaction events

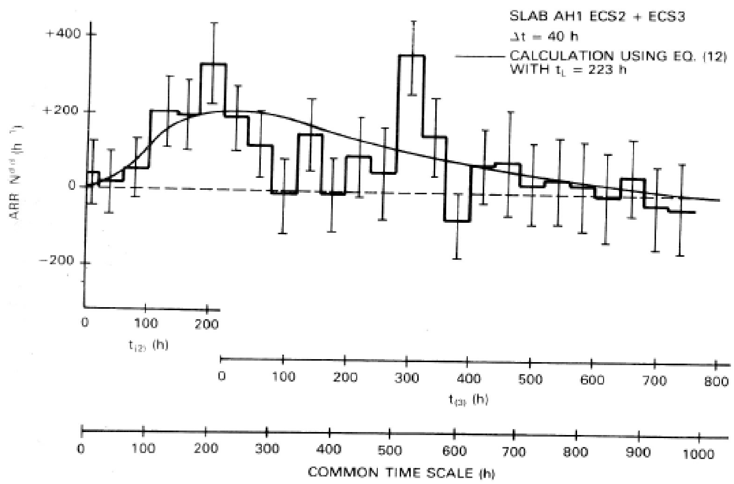


Fig. 6: Average reaction rate in 20h time bins for electrode AH1 compared to the curve from the plasma-like reaction model /9/; the experimental data are both from ECS2 and ECS3

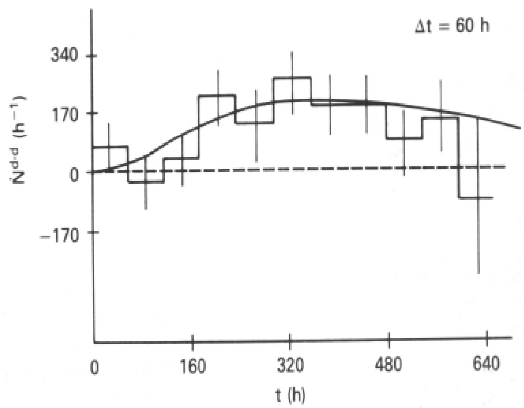


Fig. 7: Average reaction rate in 60h time bins for electrode Z1 compared to the model curve /6/

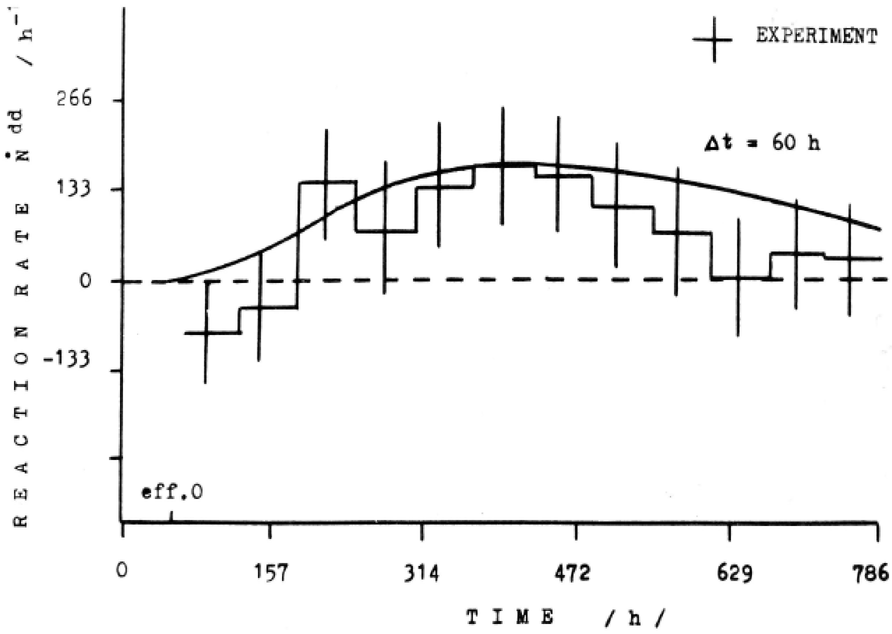


Fig. 8: Average reaction rates for electrode Z2 in 60h time bins /8/ compared to the curve calculated by the plasma-like model

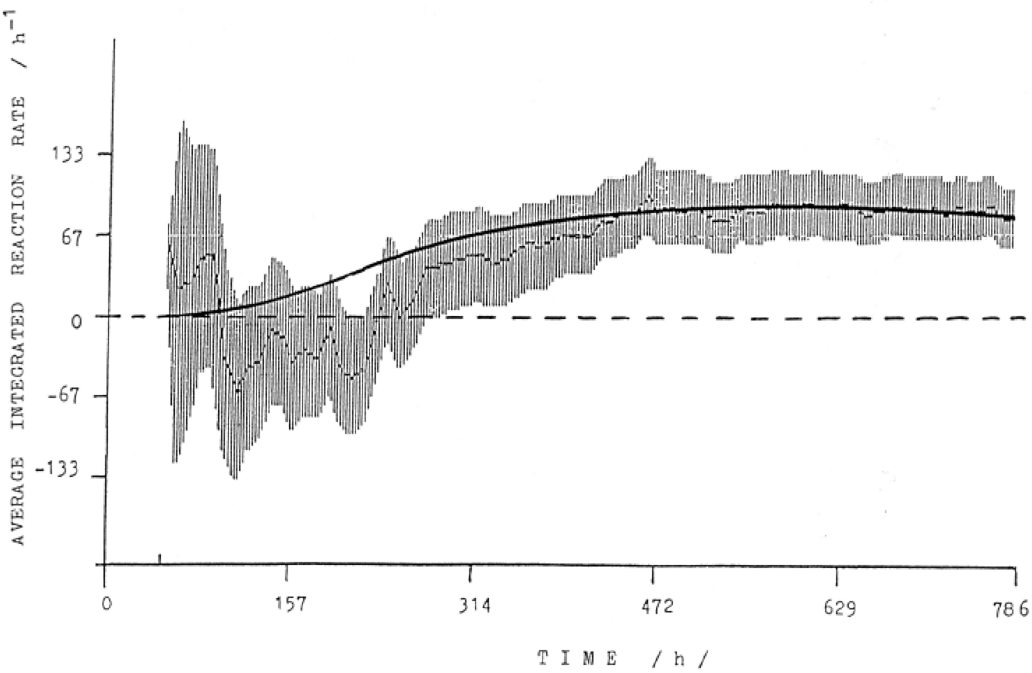


Fig. 9: Average integrated reaction rate for elecztrode Z2 /8/ over the whole time of electrolytic charging in ECS3 compared to the prediction of the plasma-like model

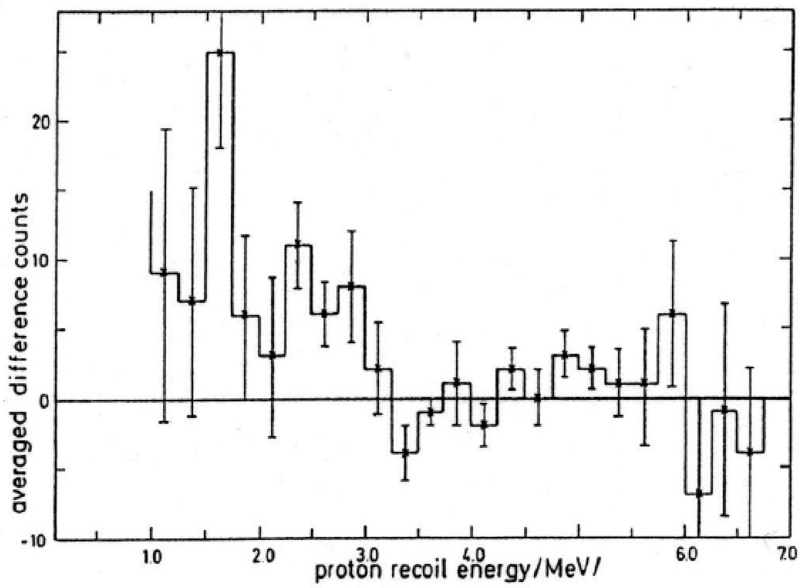


Fig. 10: Recoil proton energy spectrum from absorpion of Deuterium is the gas phase in Titanium turnings [10]

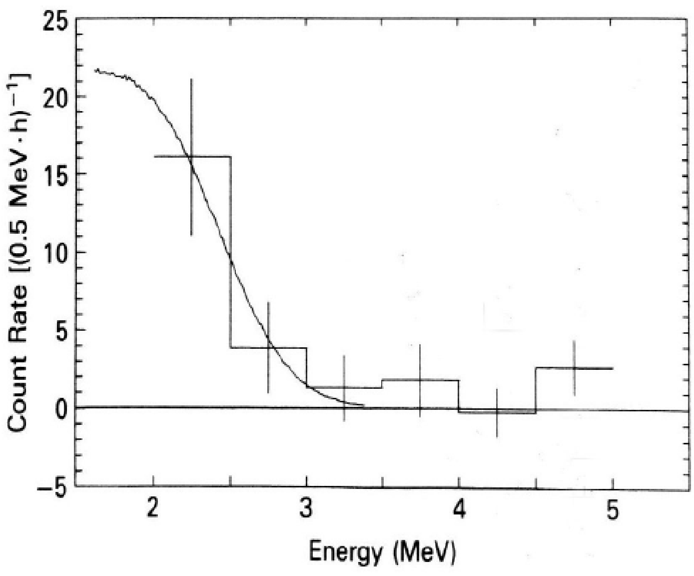


Fig. 11: Recoil proton energy spectrum measured during degassing of a massive Palladium cylinder compared to the dd-spectrum measured at a neutron genertor [11]

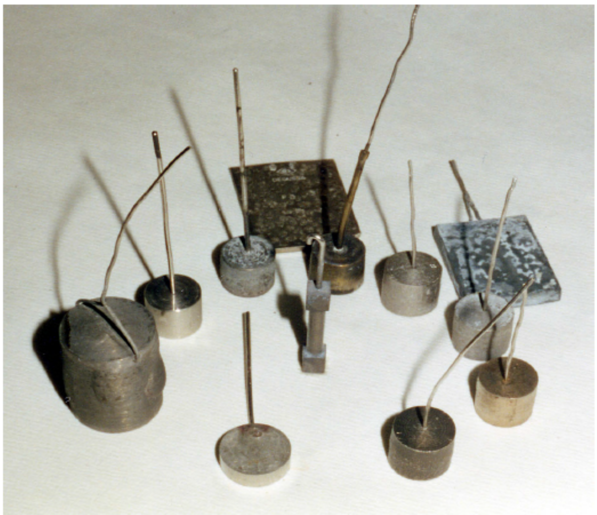
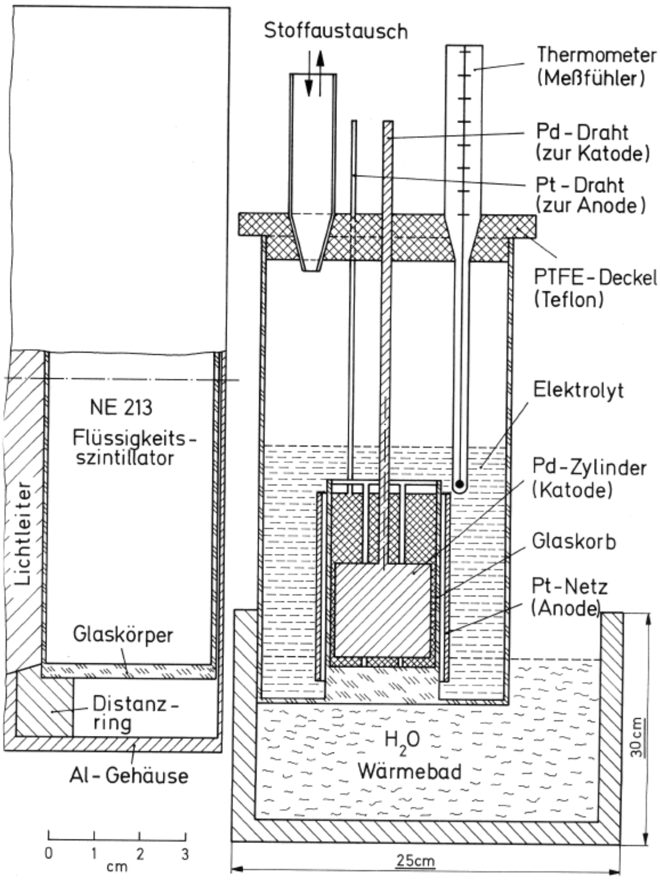
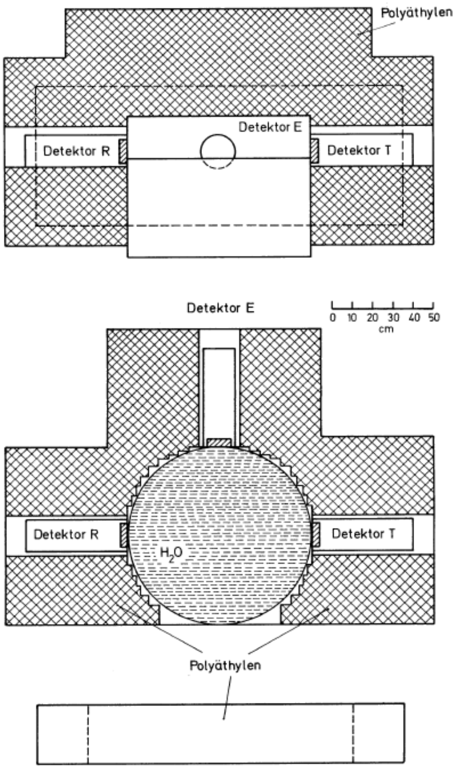


Fig. 12: Foto of all massive Palladium cathodes used for investigations at TUD

Fig. 13: Drawing of electrolytic cells used for both neutron and Tritium Measurements; experimental arrangement as used in ECS2-4 (right)





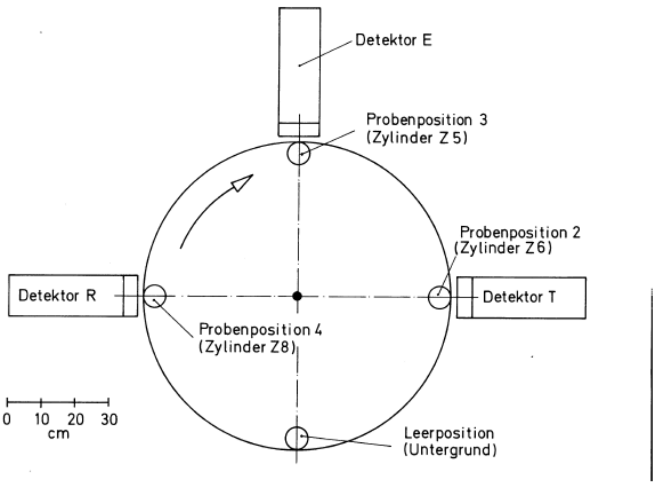


Fig. 14: schematic drawing of the ECS5 experimental arrangement using three detectors R, E, T and three electrolytic cells with cathodes Z5, Z6 and Z8 located in a water bath, that can be turned around

Fig. 15: same arrangement as on fig. 13, including heavy polyethylene shielding (left)

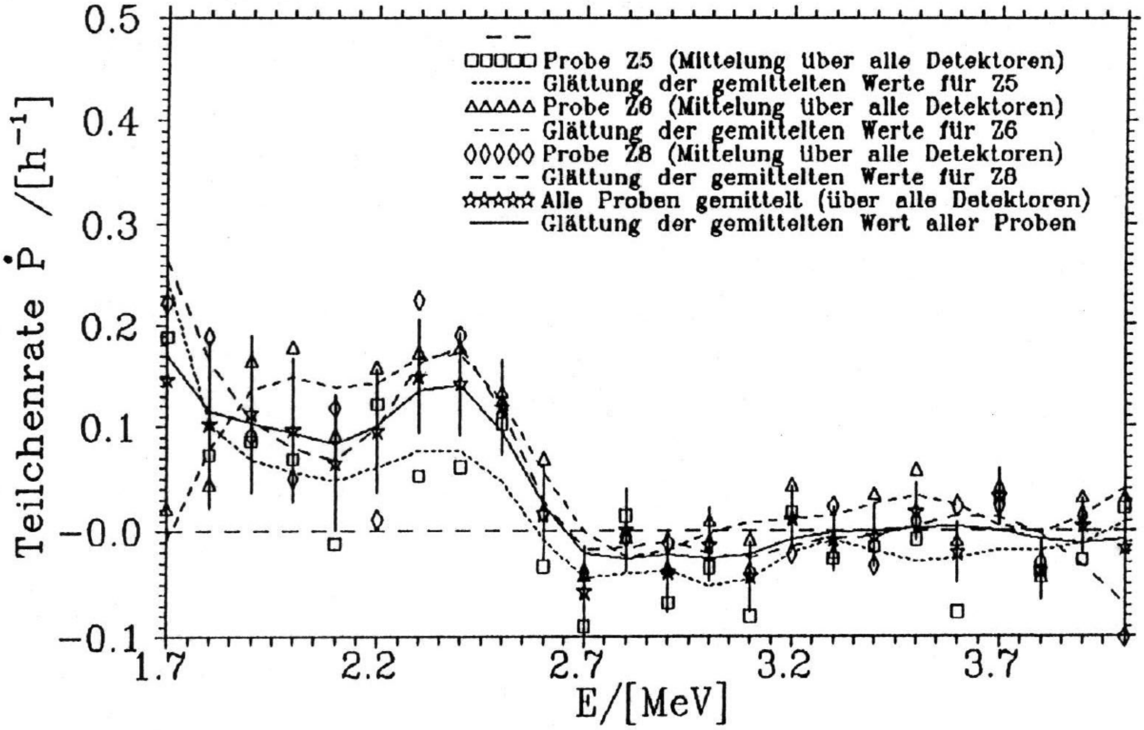
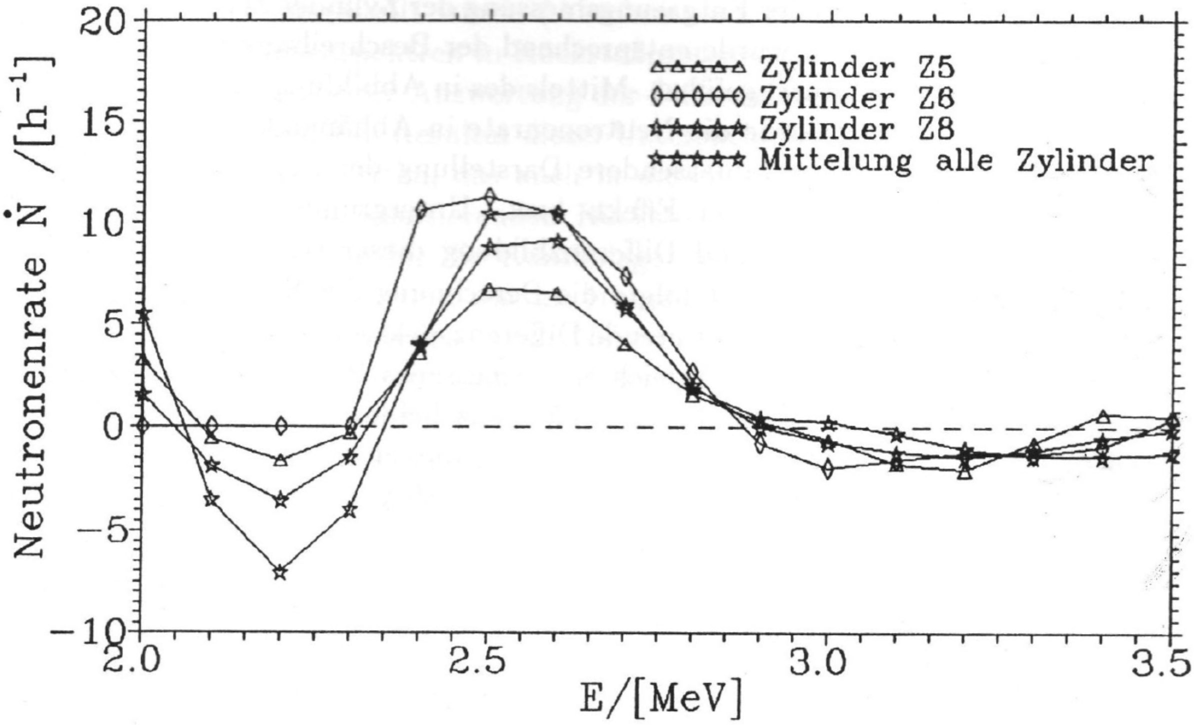


***Fig. 16:** Experimental recoil proton spectra obtained for different cathodes Z5, Z6 and Z8 and averaged spectra for three cathodes from ECS5 meadurements*

***Fig. 17:** Neutron spectra obtained from unfolding the recoil proton spectra for different cathodes and for averaged spectrum for all three cathodes from ECS5 measurements*



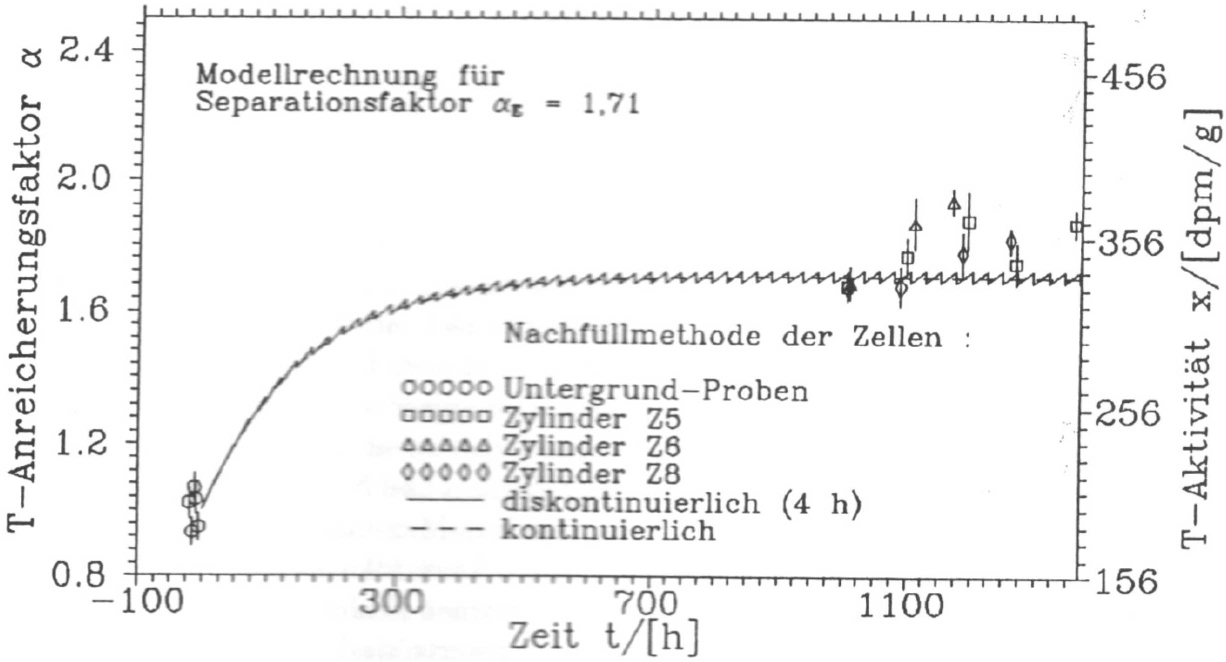


Fig. 18: Experimental data of Tritium measurements within ECS5 compared to a model calculation showing the enrichement factor α continuos and discontinous refilling of D_2O losses by electrolyses; after about 800 h of electrolysis a saturation level is reached, pending on the Tritium production rate; lower data at left side – fresh electrolyte; upper data right side - Tritium data for cells Z5, Z6, Z8 and average after more than 1000 h of electrolysis; the graph shows a definite increase of Tritium activity due-to rection processes in the samples