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Analysis of energy spectrum of neutrons in cold-fusion experiments by the TNCF model

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Summary. The energy spectra of neutrons in the cold-fusion phenomenon measured by Bressani *et al.* in a Ti/D gas loading system were analyzed with the TNCF model proposed by one of the authors (H. Kozima). The result shows that the data of positive results are interpreted consistently with a value of the adjustable parameter $n_n = 10^4\ 10^7\ \mathrm{cm}^{-3}$. These values of n_n are in the smallest range of values determined hitherto in various materials used in the cold-fusion experiments with positive results. A possible cause of the small value of the parameter n_n is discussed taking into consideration the characteristics of the sample.

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1. – Introduction

Precise measurements of the energy spectrum of neutrons emitted from a cold-fusion (CF) system have been performed by Bressani *et al.* [1-3] soon after the discovery [4] and confirmation [5-7] of the cold-fusion phenomenon. Moreover, the TNCF (Trapped Neutron Catalyzed Fusion) model [8-12] proposed by one of the present authors (H. Kozima) has been applied successfully in these five years to various events of the cold-fusion (CF) phenomenon in which an explanation is included of such a large ratio of numbers of helium 4 and neutron, N_{He}/N_n, and of tritium and neutron, N_t/N_n, as large as 10^7 obtained in experiments by theoretical values of N_{He}/N_n and N_t/N_n of about 1×10^6.

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It was, however, postponed to apply the model for the energy spectrum of neutrons observed in several cold-fusion systems until its final completion three years ago. The model has been applied [13,14] recently to the first measurement of the energy spectrum of neutrons and to a null result in electrolytic systems. In this paper, we analyze the precise measurements [1-3] of the neutron spectrum in Ti/D systems with gas loading.

It should be noticed, however, that the long process of a reaction chain from the trigger reactions to the energy spectrum of neutrons in the TNCF model makes the result of this analysis rather ambiguous compared with those analyses applied to more than 50 events in experimental results hitherto and given in previous papers [11,12,15] and in a book [16]. Neverthless, the analysis given in this paper will show the reality of the (CF) phenomenon through the appropriate explanation of the precise measurements consistent with many other data and will also show the effectiveness of a phenomenological approach.

2. – Experimental results of Bressani *et al.*

Bressani *et al.* [1-3] have made precise measurements of energy spectra of neutrons emitted from gas-loaded Ti/D systems. Their measurements were performed with the time-of-flight method with double-scattering technique to measure neutron energy and consisted of three sets of metals with Ti shavings (A), with Ti sponge (B) and with Ti metallic shaving and sponge (C) with measurements of the loading ratio $X \equiv$ D/Ti as introduced as follows.

Set A. *Experiment with* Ti *shaving sample* [1].

In the first set of their experiment a hydrogen isotope gas with a pressure up to 1.5×10^3 Torr was used to load deuterium or protium into Ti metal (3 g) with a shape of shaving. Neutron emission was measured in thermocycles between 25 and 540 °C.

A small enhancement of the events around 2.45 MeV (2–3 MeV) was observed with D_2 gas but not with H_2 gas. The background in the neutron energy spectra was totally instrumental, inherent to the technique, *i.e.* due to the photomultipliers' noise. A clear peak centered at ~ 2.5 MeV was visible with a satisfactory background subtraction around the peak compatible with that expected by a Monte Carlo simulation.

The neutron emission with energies between 2 and 3 MeV measured in this experiment was 4.0 ± 1.5 n/s corresponding to 1.3 ± 0.5 n/s $\cdot$ gTi without burst-type emission. The statistical significance of the 2.45 MeV neutron emission was $\sim 2.5\,\sigma$. The mean (over the sample) loading ratio D/Ti, X, was 0.32.

Set B. *Experiment with* Ti *sponge sample* [2].

In the second set of their experiment with Ti and Pd metals, better control of the pressure and temperature of both the metal and the gas had been performed which made the statistical significance larger. For the Ti experiment, high-purity Ti sponge (20 g) was used. During the repeated thermocycles between 25 and 540 °C, the morphology of the Ti sample gradually changed from sponge to a powder due to the large strains associated with the hydrides formation and phase transformations. As a background measurement, several cycles were performed by filling the sample cell with protium instead of deuterium.

An estimate of the neutron emission between 2 and 3 MeV per unit mass and time was made assuming that the neutron production rate was independent of time and gave 0.11 ± 0.03 n/s $\cdot$ gTi. There was no neutron burst at all. The statistical significance was $5\,\sigma$. The mean D/Ti ratios of samples at start, X_0, were 0.7 and 1.8.

For Pd/D instead of the Ti/D system, they observed a 2.45 MeV neutron emission rate of 0.02 ± 0.01 n/s $\cdot$ gPd without any burst, too.

In set B, a more complete experiment than set A, the presence of neutrons with energies up to 7 MeV was apparent in the spectra with a lower statistical significance (2–3 σ) than those of $\sim$ 2.5 MeV with $\sim$ 5 σ. In this experiment, by the author's opinion expressed in the recent paper [3], the neutron hodoscope was optimized for the detection of 2.5 MeV neutrons, and then the results on the presence of higher-energy neutrons could be biased.

Set C. *Experiment with* Ti *metallic shaving and sponge samples* [3].

Due to the possible presence of neutrons at energies higher than 2.5 MeV, the energy range of detected neutrons in this set C, with an experimental arrangement very similar to but up-graded with respect to that in set B, was extended up to 8 MeV.

Two cases out of six, depicted in their fig. 4 (with metallic shaving sample, C1) and fig. 8 (with sponge sample, C2) in the paper [3], have shown a weak emission of neutrons of the order of 0.10 neutrons $g^{-1}s^{-1}$ at a 2–3 σ level achieved without clear correlation with the initial loading ratio X_0. The energy spectra of the former (C1) peaked in the energy range 2–2.8 MeV and of the latter (C2) distributed widely from 0.4 to 7.6 MeV. In these cases, the net flux of neutrons are 0.10 ± 0.04 and 0.10 ± 0.05 neutrons $g^{-1}s^{-1}$ and the initial loading ratios X_0 were 1.125 (C1) and 1.837 (C2), respectively.

The experimental results in set C show clearly irreproducibility, or rather qualitative reproducibility, of the CF phenomenon, neutron emission in this case. Uncontrollable minor differences in the experimental procedure result in finite differences from null results to positive results with various shapes of the energy spectrum.

The experimental data of these three sets with positive results of measurements A (with Ti shaving), B (with Ti sponge) and C (with Ti metallic shaving and sponge) on the Ti/D system can be summarized as follows: the 2.5 MeV neutrons are emitted from a Ti/D system with the statistical significance of 2.5 σ (A), 5 σ (B) and 2–3 σ (C1). The shape of the neutron spectrum was slightly different in the three sets. In one case in set C (C2), a spectrum distributed from 0.4 to 7.6 MeV was observed.

The mass emission rate in set A and in sets B and C was different by one order of magnitude: 1.3 ± 0.5 (A), 0.11 ± 0.03 (B), 0.10 ± 0.04 (C1) and 0.10 ± 0.05 n/s $\cdot$ gTi (C2). These values of the emission rate are used to calculate the parameter n_n of the TNCF model in the next section. It is noticed that the energy spectrum in set C2 was broad and only neutrons contributing to an energy range 2–3 MeV (say one-forth of the total) should be taken in the calculation to compare with others.

The difference of neutron emissions in set A and set B was interpreted by the authors [2] to indicate that the CF phenomenon in this system is essentially a bulk and not a surface process because the surface-to-volume ratio S/V is larger in Ti sponge (B) than in Ti shaving (A).

We analyze the above data on the TNCF model in the next section.

3. – Analysis of the data by the TNCF model

In the CF system with gas loading of hydrogen isotopes, there is no metal layer on the surface of the solid sample and the important role of trigger reactions played in the metal layer in electrolytic systems is missing. The relevant reactions in this case are written down as follows in the TNCF model with cross-sections given in data books [17, 18].

The trigger reaction in the Ti/D system is that between a trapped neutron and the occluded deuteron in volume:

$$\text{n} + \text{d} = \text{t}\,(6.98\,\text{keV}) + \gamma\,(6.25\,\text{MeV}). \tag{1}$$

The cross-section of this reaction is 5.5×10^{-4} b (1 b $= 10^{-24}\,\text{cm}^2$) for a thermal neutron.

The triton with energy $\varepsilon = 6.98$ keV generated in this trigger reaction can induce a succeeding breeding reaction with a deuteron, the cross-section of which is 3.0×10^{-6} b in the path of length ℓ ($\sim 1\,\mu$m):

$$\text{t} + \text{d} = {}^4\text{He} + \text{n}. \tag{2}$$

The photon with energy 6.25 MeV generated in the trigger reaction (1) can induce photodisintegration of a deuteron (threshold energy ~ 2.2 MeV) giving a neutron with an energy of about 2 MeV:

$$\gamma\,(6.25\,\text{MeV}) + \text{d} = \text{n} + \text{p}. \tag{3}$$

The cross-section of this reaction is about 2 mb (2×10^{-3} b).

The neutron with an energy larger than 14.1 MeV generated in the reaction (2) can accelerate or dissociate deuterons to generate neutrons:

$$\text{n} + \text{d} = \text{n}' + \text{d}', \tag{4}$$

$$\text{n} + \text{d} = \text{n} + \text{p} + \text{n}. \tag{5}$$

The cross-sections of the elastic collision (4) and the disintegration (5) are 0.62 and 0.18 b, respectively.

The accelerated deuteron d′ (up to an energy of 12.5 MeV) in the reaction (4) can induce direct d-d reactions:

$$\text{d}' + \text{d} = {}^3\text{He} + \text{n}, \tag{6}$$

$$= \text{t} + \text{p}. \tag{7}$$

The cross-sections of the reactions (6) and (7) for a deuteron of 12.5 MeV are 8.9×10^{-3} and 3.1×10^{-3} b, respectively.

Thus, neutrons with energies between 2 and 3 MeV can be generated in several reactions in the chain from (2) to (6) in the TNCF model. To analyze the energy spectrum of neutrons from our point of view, therefore, it is necessary to calculate the number of neutrons generated in reactions (3), (4), (5) and (6). In this paper, we confine relevant reactions only to those given above although there are evidences showing the importance of reactions between thermal neutrons and lattice nuclei in the sample [15].

The rate P_f of the reaction (1) per unit time is expressed by the following relation:

$$P_f = 0.35\, n_{\text{n}}\, v_{\text{n}}\, n_{\text{d}} V \sigma_{\text{nd}} \xi, \tag{8}$$

where $0.35\, n_{\text{n}} v_{\text{n}}$ is the flow density of the thermal neutron per unit area and time, n_{d} is the density of the deuterons in the reaction region with volume V and σ_{nd} is the cross-section of the reaction. In the following analysis, we take V as volume of the sample. The factor ξ expresses an order of instability of the trapped neutron in the reaction space:

TABLE I. *Values of the adjustable parameter $n_{\rm n}$ for the sets A, B and C.*

Set	A ($X = 0.32$)	B ($X_0 = 1.8$)	B ($\overline{X}_0 = 1.25$)
$n_{\rm n}$ (cm^{-3})	3.8×10^7	5.5×10^4	2.7×10^4
Set	C1 ($X_0 = 1.125$)	C2 ($X_0 = 1.837$)	
$n_{\rm n}$ (cm^{-3})	4.4×10^4	1.4×10^4	

we take $\xi = 0.01$ for reactions which occur in the volume (and $\xi = 1$ for reactions in the surface layer) according to the recipe of the TNCF model [11, 15, 16].

The energetic particles generated by the trigger reactions react with particles in the crystal lattice and cause the breeding reactions (2) to (7). The rate of reactions per unit time, $P_{\rm r}$, between an energetic particle with energy ε and one of stable nuclei in the solid is given by a formula similar to that in vacuum:

$$P_{\rm r} = N_\varepsilon n_{\rm N} \sigma_{\rm N} \ell, \tag{9}$$

where N_ε is the number of particles with energy ε generated in the sample per unit time, ℓ is the path length of the energetic particle (taken as 1 μm for a charged particle and as the size of the sample for a photon in the following calculation), $n_{\rm N}$ is the density of the nucleus, $\sigma_{\rm N}$ is the cross-section of the reaction.

Applying these relations (with $\xi = 0.01$ in (8)) to the experimental data explained in sect. **2** as done in a previous paper [12], we obtain the values of $n_{\rm n}$ shown in table I for the sets A, B and C (with value of loading ratio $X \equiv$ D/Ti in the bracket).

In this calculation, the experimental value of neutrons with energies between 2 and 3 MeV (summarized at the end of sect. **2**) was assumed as the number of neutrons generated in the overwhelmingly predominant reaction (3) (because the contribution of reactions (5) and (6) to neutrons in this energy range is negligible). To calculate $n_{\rm d}$ in eq. (8), the average or initial D/Ti ratios were used. In the case of set B, the maximum value $X_0 = 1.8$ and the average value $\overline{X}_0 = 1.25$ (of 0.7 and 1.8) are used, for illustration. These values of $n_{\rm n}$ determined with those assumptions explained above are in the range of the smallest values determined before but not inconsistent with them [10, 11, 16].

On the other hand, the neutron with an energy of 14.1 MeV generated in the reaction (2) will contribute to the whole range of the energy spectrum up to 14.1 MeV lowering its energy and generating neutrons by such reactions as (4) and (5). (n, 2n) and (n, 3n) reactions of the neutron with one of the lattice nuclei be included in this case.

A result of numerical calculation [14] of the energy spectrum of neutrons generated in the Pd/D/Li system is shown in fig. 1 for an illustration. In this case, the trigger reaction is between the thermal neutron and $^6_3$Li:

$$\mathrm{n} + {}^6_3\mathrm{Li} = {}^4_2\mathrm{He}\,(2.1\,\mathrm{MeV}) + \mathrm{t}\,(2.7\,\mathrm{MeV}). \tag{10}$$

The high-energy triton produced in this reaction generates higher-energy particles than that in the reaction (1) and the succeeding reactions differ very much from those considered above although the characteristic of the spectrum not confined to around 2.5 MeV should be common.

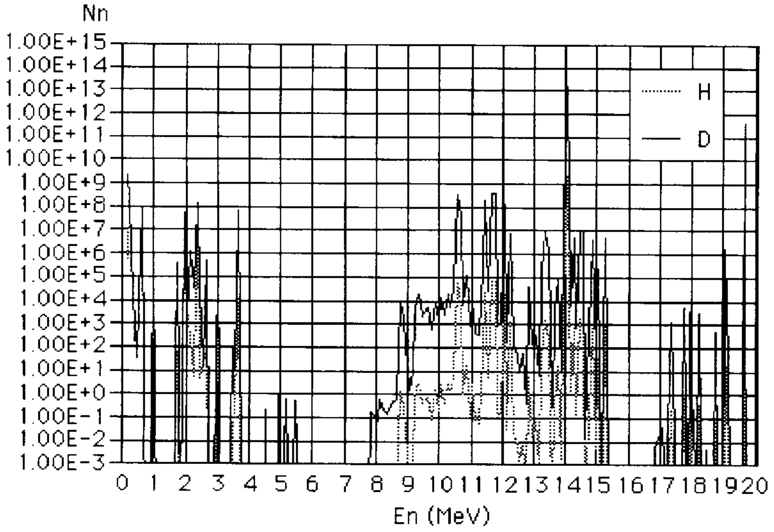

Fig. 1. Numerical result of the energy spectrum of neutrons generated in Pd/D/Li and Pd/H/Li systems by the TNCF model. The simplifications were made that the PdD/Li (PdH/Li) layers have a thickness of 1 μm on the surface of Pd metal where neutrons with $n_n = 10^{10}\,\mathrm{cm}^{-3}$ are trapped and that Pd lattice is treated as a continuous medium where energetic charged particles have flight lengths of 1 μm in it.

It is seen in this figure that there are neutrons with energies from about 2 MeV up to about 15 MeV. Essentially the same figure is obtained for the Ti/D system with a larger value of n_n than that used in this calculation. This figure reproduces qualitatively a characteristic of the spectrum extending to energies higher than 2.5 MeV as observed in the experiments [1-3] in the Ti/D system and in other experiments [19] and is consistent with them, especially with that of C2. This consistence gives an evidence of the fundamental validity of the guiding principles assumed in the TNCF model.

4. – Discussion

The TNCF model was applied to the energy spectrum of neutrons emitted from the gas loading Ti/D system. The model with a single adjustable parameter n_n and based on the conventional physics had been proposed to understand systematically and consistently the whole bulk of experiments in the CF phenomenon characterized by qualitative reproducibility occurring in the complex system, composed mainly of transition metal hydrides and deuterides with some other undetermined characteristics. The result obtained in this paper shows semi-quantitative explanation of the spectra obtained by the most precise and reliable measurements done in this field.

The parameters n_n in the TNCF model of 10^4 $10^7\,\mathrm{cm}^{-3}$ determined above for the data sets A, B and C are rather small compared with those values of 10^8 $10^{12}\,\mathrm{cm}^{-3}$ determined in previous analyses [10, 11, 16] of other experiments in the CF phenomenon. These values of n_n determined in this work, however, should be taken within an error of one or two orders of magnitude due to uncertainty in the experimental values used in the calculation, *e.g.*, n_d changes in the process of the thermocycle employed in the experiments as explained in the paper [3].

As a cause of the discrepancy between the values obtained in this paper and those of previous analyses, it is possible to consider that the parameter ξ is smaller in the TiD_x system than 0.01 determined in a Ni/H/K system according to a result of our analysis [20]. The small number of analyzed data [21] for samples without surface layer of alkali metals gives uncertainty in the value of ξ itself, and the final determination of its value in and its dependence upon the material should be considered as pending.

Another cause of small values of n_n determined in this work might due to the lack of the surface layer made of alkali metals, essential to trap neutrons in the sample in the case of electrolytic systems.

The establishment of necessary conditions for neutron trapping is accomplished by stochastic atomic processes in the sample and is related closely with the problem of the irreproducibility of the CF phenomenon. In the second paper [2], the authors cited a paper with a null result in a similar Ti/D system to theirs and commented the probable origin of the difference due to the difference of the thermocycles used in one and the other experiments. They have also reported null results in four out of six runs in their third paper [3], as explained above in sect. **2**.

From our point of view, the differences of shape and quality of the sample and of the experimental processes (temperature range and temporal period of thermocycles, quality of the sample, and so on) influence the condition for neutron trapping and for the rates of trigger and breeding reactions through the difference in deuteron distribution and surface and boundary structures of the sample. In the gas loading system, these experimental processes influence decisively the distribution of hydrogen isotopes in the metal, which is sensitive to the trapping of neutrons and the reactions between particles in the sample. Thus, the CF phenomenon is inevitably irreproducible, or is characterized by qualitative reproducibility, inherent in the stochastic processes of atomic motion in solids yielding from null to positive results with various shapes of energy spectrum.

The widely distributed energy spectrum from 0.4 to 7.6 MeV in set C2 should be explained by whole nuclear reactions of trapped neutrons with other nuclei in the material, especially with those in boundary regions, whose existence was recognized in recent data of the nuclear transmutation in solids [15, 16].

The absence of neutron emission in the Ti/H system used in set A [1] might be, from our viewpoint, a result of an expected reaction in the system with a fairly large cross-section of 0.332 b;

$$\text{(11)} \qquad \mathrm{n} + \mathrm{p} = \mathrm{d}\,(1.33\,\mathrm{keV}) + \gamma\,(2.22\,\mathrm{MeV}),$$

the products of which are inactive to induce reactions like (3) generating a neutron with an energy of ~ 2 MeV even if comparable values of the parameter n_n are realized in both systems.

Thus, one of the fundamental questions in the CF phenomenon about the possibility of d-d reactions in solid at room temperature was overcome by the existence of energetic deuterons produced in or accelerated by energetic particles generated in trigger and breeding reactions in the TNCF model. However, the number of neutrons with energies of 2.45 MeV and of more than 3 MeV is very small compared with those of ${}^4_2\mathrm{He}$, t and transmuted nuclei in appropriate systems, as recognized recently.

The physical basis of the TNCF model has been investigated [15, 22-24] on the nuclear physics and solid-state physics, showing a possible step to construct solid-state nuclear physics where neutrons in the solids play a fundamental role to realize the whole phenomenology of the CF phenomenon from production of excess heat to nuclear transmutation.

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