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#### **CFP** (**Cold Fusion Phenomenon**) stands for

"Nuclear reactions and accompanying events occurring in open (with external particle and energy supply), non-equilibrium system composed of solids with high densities of hydrogen isotopes (H and/or D) in ambient radiation" belonging to Solid-State Nuclear Physics (SSNP) or Condensed Matter Nuclear Science (CMNS).

This is the *CFRL News* (in English) No.93 for Cold Fusion researchers published by Dr. H. Kozima, now at the Cold Fusion Research Laboratory, Shizuoka, Japan.

This issue contains the following items:

- 1. From the History of CF Research (7) The First Reliable Measurement of Tritium in Pd/D/Li System by Packham et al. (1989)
- 2. H. Kozima, "The TNCF Model and the E-Cat" was published in E-Cat World website.
- 3. French Cold Fusion Research Group was established
- 1. From the History of CF Research (7) The First Reliable Measurement of Tritium in Pd/D/Li System by Packham et al. (1989)

This series "From the History of CF Research" reached the observation of tritium, one of the direct evidences of the CFP, which is a controversial theme between pros and cons of this phenomenon. Before the discussion of this problem, we list up below the past and present themes discussed in this series:

- (1) DOE Report 1989 and DOE Report 2004, *CFRL News* No. 87 (September 1, 2014)).
- (2) Detection of  ${}^4_2$ He by Morrey et al. (1990), *CFRL News* No. 88 (November 20, 2014).
- (3) The First Observation of Nuclear Transmutation in a Protium System by R.T. Bush and R.D. Eagleton (1993, 1994), *CFRL News* No. 89 (January 10, 2015).
- (4) The First Measurement of the Energy Spectrum of Neutrons emitted in the CFP by S.E. Jones et al. (1989), *CFRL News* No. 90 (February 10, 2015).
- (5) The Most Extensive Measurement of Excess Energy by M.C.N. McKubre et al. (1993, 1994), *CFRL News* No. 91 (March 10, 2015).
- (6) The First Confirmation of Localized Nuclear Reactions in CFP by M. Okamoto et al. (1994), *CFRL News* No. 92 (April 10, 2015).
- (7) The First Reliable Measurement of Tritium in Pd/D/Li System by Packham et al. (1989), *CFRL News* No. 93 (May 10, 2015).

#### 1.1 Introduction

The discovery of the cold fusion phenomenon (CFP) had been announced by Fleischmann and Pons in 1989. Their paper, published in *J. Electroanal. Chemistry*, had reported measurements of excess heat, gamma ray spectrum, neutron flux, tritium generation/accumulation in a Pd/D/Li system [Fleischmann 1989].

According to our classification of experimental data into two categories, *Direct and Indirect Evidences* for the nuclear reaction in the CFP, the Direct Evidence is useful to identify the character of events [Kozima 1989 (Chap. 6), 2006 (Sec. 2.2.1.1)]. The same consideration had been recognized by many researchers as expressed as "It is the contention of the authors that the alleged phenomenon is better characterized by the production of nuclear particles than by the measurement of bursts of heat."[Packham 1989].

In the direct evidences, tritium and helium-4 ( ${}^{4}_{2}$ He) have been main targets of the researches of the cold fusion phenomenon from the first. And then, transmuted nuclei had been recognized as one of the important direct evidences.

The situation outlined above is understood well if we recollect the presumption supposed by the pioneering researchers. They assumed occurrence of the d-d fusion reactions in CF materials (e.g.  $PdD_x$ ) at around room temperature which are written down as follows (in free space):

$$d + d \rightarrow {}^{4}_{2}\text{He}^{*} \rightarrow t (1.01) + p (3.12), \qquad Q = 4.13$$
 (1.1)  
  $\rightarrow {}^{3}_{2}\text{He} (0.82) + n (2.45), \qquad Q = 3.27$  (1.2)

$$\rightarrow$$
 <sup>4</sup><sub>2</sub>He (0.08) +  $\gamma$  (23.8),  $Q = 23.8$  (1.3)

The branching ratios of these fusion reactions are determined in nuclear physics as

$$1:1:10^{-7} \tag{1.4}$$

in free space at an energy range up to several MeV of initial deuterons and supposed to be correct down to the thermal energy range.

Assuming that the branching ratios given above are applicable to the cold fusion phenomenon (CFP), we can deduce several conclusions about the measurement of tritium (t), proton (p), very difficult), helium-3  $\binom{3}{2}$ He), neutron (n), helium-4  $\binom{4}{2}$ He) and gamma (y). If the CFP is induced fundamentally by the d-d fusion reactions, then we can expect relations given in Eq. (1.4) between numbers  $N_x$  of measured particles x's, where x stands for t, (p), He-3, n, He-4, y or Q (excess energy), as follows:

$$N_{\rm t} = (N_{\rm p}) = N_{\rm He-3} = N_{\rm n} = 10^7 N_{\rm He-4} = 10^7 N_{\gamma} \approx N_Q.$$
 (1.5)

It should be noticed that the number of reactions  $N_Q$  for the excess energy Q is defined as  $N_Q = Q$  (MeV)/7.4 (MeV) where 7.4 MeV is the total excess energy liberated by the reactions (1.1) and (1.2). In this case, we expect to measure tritium, (proton), helium-3 and neutron with the same probability but no helium-4 and gamma.

On the other hand, if the reaction (1.3) occurs predominantly (almost 100%) in contradiction to the ordinary branching ratios known in nuclear physics, by any chance with a multi-phonon emission instead of a gamma, the relations (1.5) are replaced by the following one:

$$N_{\text{He-4}} = N_{O'}$$
, and  $N_{\text{t}} = (N_{\text{p}}) = N_{\text{He-3}} = N_{\text{n}} = 0$ . (1.6)

In this relation  $N_{Q'}$  expresses the number of reactions producing the excess energy Q' thermalized in the CF material (e.g.  $PdD_x$ ) and is defined as  $N_{Q'} = Q'$  (MeV)/23.8 (MeV). In this case, we expect to measure helium-4 in accordance with the excess energy 23.8 MeV for an atom of  $^4_2$ He but no tritium, (no proton), no helium-3 and no neutron.

It should be emphasized that the Eq. (1.5) and the Eqs. (1.6) contradict each other. Furthermore, the Eq. (1.5) for the occurrence of d-d fusion reactions (1.1)-(1.3) (or that the Eqs. (1.6) for the modified d-d fusion reaction (1.3) by any chance) is the litmus test to determine whether the presumption that the d-d fusion reaction(s) assumed as the fundamental mechanism for the CFP in deuterium-occluding CF materials is right or not. Once tritium is measured in an experiment, then the relations (1.6) is denied even if the amount of the observed tritium is not accordance with the other observables (e.g. neutron) denying the relation (1.5). If the relation (1.5) (or relations (1.6)) is denied by experiments, then we have to look for other reactions than the d-d fusion reactions responsible to the various events observed in the CFP.

It is suggestive to recollect the comment by Fleischmann in his historical paper in relation to this discussion:

"The most surprising feature of our results however, is that reactions (v) and (vi) are only a small part of the overall reaction scheme and that the bulk of the energy release is due to an hitherto unknown nuclear process or processes (presumably again due to deuterons)."[Fleischmann 1989 (Discussion)]

Reactions (v) and (vi) referred in above citation correspond to Eqs. (1.1) and (1.2) in this paper. Fleischmann's intuition might be able to sense the subtle difference between the experimental result he obtained and the framework of the d-d fusion reactions.

In this paper, we will investigate the experimental data sets of tritium measurements in terms of the interest explained in the previous paragraph.

#### 1.2 Experimental Data Sets by Pioneering Researchers

By May, 1989, the *Energy Research Advisory Board* to the United States Department of Energy (DOE) was asked to "Review the experiments and theory of the recent work on cold fusion." The Board submitted their Report to DOE on November, 1989 [DOE Report 1989] in which many experts on the relevant fields of science to the cold fusion phenomenon contributed to investigate extensively scientific value of "the apparent observations of cold fusion and significant quantities of energy from these phenomena" since April to this time.

The most reliable measurements of tritium in the early stage of investigation are pointed out as Packham et al. [Packham 1989], Wolf et al. [Wolf 1989] and Iyengar et al. [Iyengar 1989] among others in the [DOE Report 1989].

We summarize the data obtained in the early stage of the CF research including the papers referred above and give DOE evaluation on some of them in this Section.

The tritium measurements have been performed using several types of CF materials; (1) Pd/D/Li and PdSi<sub>x</sub>/D/Na electrolytic systems. The most popular one is Pd metals hydrogenated by electrolysis with electrolyte D<sub>2</sub>O + LiOD. This type of CF material (Pd/D/Li) was used by Fleischmann at al. [Fleischmann 1989], Packham et al. [Packham 1989], Wolf et al. [Wolf 1989], Martin [Martin 1989], Iyengar et al. [Iyengar 1989 (Sec. 5)], Storms et al. [Storms 1990], Chien et al. [Chien 1992], Iwamura et al. [Iwamura 1994]. On the other hand, PdSi<sub>x</sub>/D/Na system was used by Iyengar et al. [Iyengar 1989] and Claytor et al. [Claytor 1993, Tuggle 1994].

#### (1) Pd/D/Li and PdSi<sub>x</sub>/D/Na Electrolytic Systems.

Many measurements of tritium have been performed with Pd/D/Li system and one by Iyengar et al. with PdSi<sub>x</sub>/D/Na system. Typical examples of them are cited above. The first reliable measurements by Packham et al. [Packham 1989] were performed with this system. They observed tritium at levels  $10^2 - 10^5$  times above that expected from the normal isotopic enrichment of electrolysis in a system  $D_2O + 0.1$  M LiOD electrolysis with Pd cathode and Ni gauze anode (Pd/D/Li system).

In the experiment by Wolf et al. [Wolf 1989] in Texas A&M University performed at the same period to the above one, they observed neutron and tritium. The observed neutron emission was with a rate of 3-4 times the back-ground rate of 0.8 n/min. The tritium was determined several days after the neutron–production runs as  $5 \times 10^{12}$  tritium atoms in the solution of electrolytic cell.

In the experiment by Iyengar et al. in BARC, India, they observed tritium and neutron in systems with various types of Pd cathodes (Pd/D/Li systems) or with PdSi alloy cathodes (PdSi<sub>x</sub>/D/Na systems) and Ni or Pt anodes [Iyengar 1989 (Secs. 3 and 5)]. After a neutron burst in the experiment with a cathode of cylindrical Pd pellet 11 mm dia. × 11.2 mm height and with an anode of Pt gauze, tritium level had shown an eight fold increase and the decrease of the level indicated that additional tritium is continuously entering the electrolyte for many days after the sharp neutron burst. Their results as a whole had shown tritium evolution from CF materials (deuterium occluded Pd, PdSix, Ti samples with various shapes) and sometimes coincident evolution of neutron and tritium without quantitative relation between them.

#### Critique by DOE [DOE 1989]

From the experimental data sets where observed tritium and neutron simultaneously or in similar conditions, it had become clear that the number of neutrons  $N_n$  (or excess energy  $N_Q$ ) and that of tritium  $N_t$  differed by several orders of magnitude, sometimes called tritium anomaly [Kozima 1989 (Sec. 6.2), 2006 (Sec. 2.6)]. The same fact was pointed out in the DOE Report as follows:

"Wolf et al [Wolf 1989] at Texas A&M looked for neutron production in Bockris type cells. An upper limit to the production rate is 1 neutron/second, which is  $10^{-10}$  less that of the tritium production rates reported with similar cells by the Bockris group [Packham 1989]. This large discrepancy from the equal production rates for neutrons and tritons required by the branching ratio in the fusion reaction (Eqs. (1.1) - (1.3)), discussed in section II.B, is inconsistent by a factor of 10,000 to 100,000, even with the secondary neutrons that must accompany the tritons produced from nuclear fusion." [DOE 1989 (Sec, IIIE4)]

"In no case is the yield of fusion products commensurate with the claimed excess heat. In cases where tritium is reported, no secondary or primary nuclear particles are observed, ruling out the known D + D reaction as the source of tritium. The Panel concludes that the experiments reported to date do not present convincing evidence to associate the reported anomalous heat with a nuclear process." [DOE 1989 (Conclusions)].

#### (2) Ti/D<sub>2</sub> and Pd/D<sub>2</sub> Gas Loading Systems.

In the experiment by Iyengar et al. Pd samples either in the form of Pd-black powder or Johnson & Matthey Pd-Ag foils [Iyengar 1989 (Sec. 7.1)] (Ti/D<sub>2</sub> system, n and t measured). The quantity of D<sub>2</sub> absorbed could be measured from the observed pressure drop. This corresponded to  $\approx 10^{19}$  molecules of D<sub>2</sub> gas, indicating a gross (D/Ti) ratio of hardly 0.001. However it is believed that most of the absorbed D<sub>2</sub> gas is accumulated in the near surface region [Iyengar 1989 (Secs. 7.3, 7.4 and 7.5)].

In the experiment by Tuggle et al. with a Pd/D<sub>2</sub> system, (Pd/D<sub>2</sub>, Pd powder, foil 220 micron thick and wire), four types of cells have been made: those with palladium powder and silicon powder, those with palladium foil and silicon powder, those with palladium foil and silicon powder. Layers of alternating palladium disks and silicon powder were then pressed into a ceramic form at a pressure of 11.2 MPa resulting in densities of 26% and 68% of theoretical density for the palladium and silicon respectively. In (Pd-Si cells, small solid wire, pressed powder wire, plasma cells with D<sub>2</sub> are used to get tritium) an powder composed of small (0.3 to 0.5  $\mu$  m) spheres that form chains or agglomerates up to 30  $\mu$  m in dia. [Tuggle 1994]. They observed tritium with following characteristics: "The tritium output depends on currents applied to the cells. Yet, the tritium yields depend strongly on the type of Pd metal used (powder, foil and wire) and the type of experiment, powder wire, wire, plasma."

## **Explanation of Experimental Data by TNCF Model**

At the end of this section, it will be useful to point out our explanation of tritium experiments on the TNCF model summarized in our books [Kozima 1998, 2006]. In the "Sec. 6.4 Tritium" of the Discovery of the Cold Fusion Phenomenon [Kozima 1998], we introduced the works by Srinivasan et al., Storms and Talcot, Claytor et al. Iwamura et al. [Iwamura 1994], Romodanov et al., and Bockris et al. and given their explanations on our TNCF model in "Sec. 11.7 Tritium Anomaly". In the "Sec. 2.6 Tritium" of the Science of the Cold Fusion Phenomenon [Kozima 2006], we have given essential

explanation of the experimental data of tritium production and comprehensive understanding of mutual relation among several observables such as tritium, neutron and excess heat in accordance with the experimental data.

The fundamental idea of the explanation is the nuclear reactions of trapped neutrons (neutrons in the neutron band) with deuteron ( ${}^{2}_{1}H$ ) and  ${}^{6}_{3}Li$  resulting in tritium ( ${}^{3}_{1}H$ ), and helium-4 ( ${}^{4}_{2}He$ ) and tritium ( ${}^{3}_{1}H$ );

$$n + d = t + \text{phonons (6.25 MeV)},$$
 (1.7)

$$n + {}^{6}_{3}\text{Li} = {}^{4}_{2}\text{He} (2.1 \text{ MeV}) + t (2.7 \text{ MeV}),$$
 (1.8)

where the phonons in the Eq. (1.7) are supposed to be responsible to the nuclear reaction through neutrons in the neutron band coupled to the lattice by the super-nuclear interaction proposed by us [Kozima 2006 (Sec. 3.7)].

An experimental relation between the numbers of reactions  $N_x$  and  $N_y$  producing observables x and y, respectively, e.g. relations given in Eq. (1.5) for the reactions (1.1) – (1.3), had been explained by the TNCF model within a numerical factor of 3 [Kozima 2006 (Sec. 2.6), 2014 (Sec. 5)].

#### 1.3. Experimental Data Sets after the First Stage of CF Research

There are very many works on the measurement of tritium in the CFP after the first stage of CF research discussed in the previous section [Storms 2007 (Sec. 4.4.1)].

As in the cases discussed in the previous section, the observation of tritium has been obtained in deuterium systems. This essential characteristic of the tritium production in the CFP is in accordance with the interpretation on our TNCF model referred at the last paragraph of the Section 1.2 above.

It is useful to cite here a comment (by Reviewer #1) made in the DOE Report 2004 where are pointed out some characteristics of researches of the CFP [DOE Report 2004]:

"This field is 15 years old. It has been characterized by a large number of positive but internally inconsistent results, plus an even larger number of negative results refuting many of the claims."

This comment is interpreted by our formulation as (1) Internal inconsistency denies the d-d fusion reactions resulting in relation (1.5) and (1.6) and (2) Existence of negative results denies quantitative reproducibility.

It should be remembered that the DOE Report 2004 [DOE 2004] was issued to respond the proposal offered by Hagelstein et al. [Hagelstein 2004] and had a limitation imposed by the proposal that the field of the investigation was confined in the CF materials only of the deuterium system. Due to the characteristic of the proposal, the

confinement in the deuterium system, it had a limitation in its perspective as pointed out in a comment (by Reviewer #4) [DOE 2004]:

"Curiously the theories, neither Hagelstein's nor Kozima's (see Appendix A) were discussed in the paper by Hagelstein et al. [Hagelstein 2004] (at citation)."

#### 1.4 Meaning of DOE Reports on the CF Research

As I have taken up the DOE Reports at first in this series, From the History of CF Research (1), we need communication with as many scientists in other research fields as possible. The comments given by them are precious information for us to establish the science of the cold fusion phenomenon in which we have enough knowledge of experimental facts even if we may lack experience in experimental techniques and theoretical calculations developed in each specified branches of science. In these fields of expertise we lack, it is helpful to have communication with the scientists in other fields.

However, the decisive difference between CF researchers and other scientists is the motivation to research the CFP. The response given in the DOE Reports by other scientists is confined only to the problem that touched their interests but not the CFP as a whole. Therefore, there are no perspectives beyond the problems proposed to them.

They point out key problems of the CF research as follows (Review #6):

"1. The Fusion Rate miracle. 2. The Branching Ratio miracle. 3. The Concealed Nuclear Products miracle." [DOE 2004]

These problems were used only to deny the meaning of CF research without new viewpoint how to overcome the limitation of the present knowledge in nuclear physics in CF materials (solids containing a lot of hydrogen isotopes) to meet the curious experimental data obtained in this field of the CFP.

Our answers on the TNCF model to the problems have been given already in our books and papers [Kozima 1998, 2006, 2014]. The fusion rate miracle and the branching ratio miracle are the phantom appeared from the persistence in the d-d fusion reactions (1.1) - (1.3) and resolved by other nuclear reactions than these ones between a neutron and nucleus in the system. The concealed nuclear miracle is also the result of the presumption of the d-d fusion reactions (1.1) - (1.3).

There are other contradictions between theoretical results from the presumption of the d-d fusion reactions (1.1)-(1.3) and experimental results such as tritium anomaly and higher energy neutrons discussed in (4) of this series (*From the History of CF Research* (4)). As far as we persist to the d-d fusion reactions, we have to encounter inconsistency between theoretical prediction based on the presupposed reactions and

experimental results.

Another important comment in the DOE Reports on the experimental results in the CFP is on the reproducibility:

"Some experiments have reported the production of tritium with electrolytic cells. The experiments in which excess tritium is reported <u>have not been reproducible</u> by other groups. These measurements are also <u>inconsistent with the measured neutrons on the same sample</u>. Most of the experiments to date report no production of excess tritium. Additional investigations are desirable to clarify the origin of the excess tritium that is occasionally observed." [DOE 1989 (Conclusion 2)] (Underlines at citation)

The second point of the inconsistent measurements of tritium and neutron is already explained above. The first point, the reproducibility, is based on the misunderstanding of nuclear physics as we have pointed out several times [Kozima 1998 (Sec. 9.3), 2006 (Sec. 2.14)]. The nuclear reactions in a nucleus and between nuclei are all governed by probability and described by statistical laws as the simple  $\alpha$ -decay of  $^{226}_{88}$ Ra shows at hand. It should be noticed further that the atomic and molecular processes occurring in solids in forming CF materials are stochastic and co-operative resulting in complexity which has no predictability and quantitative reproducibility [Kozima 2006 (Sec. 3.8)].

#### 1.5 Conclusion

In the case of the observation of tritium again, as reviewed in this paper, show clearly reality of new events in the cold fusion phenomenon not deniable from critical point of view as reviewers of the DOE Reports expressed even if there remains a consistent explanation of whole experimental data sets.

As previewed in Introduction, an experimental data is the litmus test of a presumption which motivated the experiment. It should be concluded that the assumption of the d-d fusion reactions in CF materials motivated the pioneers is denied by experimental facts and we have to look for a mechanism (or mechanisms) responsible to various events in CF materials of protium and deuterium systems revealed by extensive experimental works.

We have proposed a phenomenological approach with an assumption of existence of neutrons in CF materials (TNCF model) [Kozima 1994, 1998, 2006, 2014]. Based on the success of the model, we have investigated quantum mechanical bases of the model, existence of the trapped neutrons and thermalization of liberated energy in nuclear reactions, and given an answer by the formation of neutron bands in such CF materials PdD<sub>x</sub> and NiH<sub>x</sub> by super-nuclear interaction between neutrons in lattice nuclei mediated by interstitial deuterons and protons. From our point of view, the experimental facts in

the CFP suggest existence of a new science and our phenomenological approach gives a glimpse of its true shape.

After 25 years of investigation of the CFP, we have to say that we are in a shroud of darkness without any light guiding us to the science of the cold fusion phenomenon at present. The enormous number of experimental data sets reporting observation of tritium in various CF materials mainly including deuterium with no consistent explanations with other observables such as neutron, helium-4 and excess energy reminds us Poincaré's words on the science:

"The man of science must work with method. Science is built up of facts, as a house is built of stones; but an accumulation of facts is no more a science than a heap of stones is a house. Most important of all, the man of science must exhibit foresight." [Poincaré 1902].

Despite of the darkness of our sight for the science of the cold fusion phenomenon, there are some trials with different motivation to cultivate new materials such as Ni-Li-H<sub>2</sub> particles at higher temperature above 1000 °C. As we commented in an essay [Kozima 2015], it is not possible to give a new insight into the science from the break-through in technology with extraordinary conditions of environment and materials.

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# 2. H. Kozima, "The TNCF Model and the E-Cat" was published in *E-Cat World* website.

E-Cat World website: www.e-catworld.com

The essay is posted at e-catworld website:

http://www.e-catworld.com/2015/04/25/the-trapped-neutron-catalyzed-fusion-model-and-e-cat-hideo-kozima/

# 3. French Cold Fusion Research Group was established

On April 10, 2015, Mathieu Valat sent a following mail to CMNS group announcing establishment of "the French Society on Condensed Matter Nuclear Science."

"Dear fellow,

I am pleased to announce on the CMNS group, the creation of the French Society on Condensed Matter Nuclear Science.

This society is affiliated to the International Society and aims to provide a French channel for conferences, publications, discussions and collaborations on the field of CMNS. It meets the Japanese, the Russian and many other local groups that want to network, share and promote to governments and public institutions the crucial research we are pursuing altogether.

After a year of effort to fulfill the administrative hurdles, gather enthusiasts and

involved scientists, formalize the tools and draft the development of this project; we are now ready to announce officially the creation of this community to the CMNS.

One month after opening registrations, we reached forty members and the corresponding google group is already getting greatly populated with networking proposals and scientific discussions. This proves the need was there.

If you read French, you can read its website at www.sfsnmc.org.

There is also a private google group, very similar to this very one.

A Facebook wall will focus on youngsters.

For information on these matters you can reach me via info@sfsnmc.org.

Finally, a poster presenting the society will be available at ICCF19 next week.

See you there!

Mathieu Valat

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