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From the History of CF Research – A Review of the Typical Papers on the Cold Fusion Phenomenon –*

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JCF16 (The 16^{th} Annual Meeting of Japan CF-research Society) was held on December 11 - 12, 2015 in Kyoto, Japan.

Abstract

The investigation of the cold fusion phenomenon (CFP) has lasted more than a quarter of a century after 1989 when a part of its vast and diverse contents was discovered by Fleischmann et al. without remarkable success in innovation of the paradigm of modern science. Recent trend of the CF research seems shifting to the application of the CFP leaving the fundamental problems how to explain this curious phenomenon consistently with other phenomena in the frame of modern physics.

We have tried to investigate the CFP in accordance with the common sense of modern science established mainly in the 20th century. To do so, we emphasized importance of communication with other scientists working in the established fields of science.

Another point we would like to emphasize is necessity to give a great regard for the typical experimental results in this field piled up in these more than 25 years. The first measurement of the energy spectrum of neutrons in a CF system (a system where occurs the CFP) was performed by S.E. Jones et al. On the detection of ${}^{4}_{2}$ He, we have to consider the work by Morrey et al. in 1990. The first data of nuclear transmutation in a protium system was obtained by R.T. Bush and R.D. Eagleton in 1993. The most extensive measurement of excess energy was performed by M.C.N. McKubre et al. in 1993 and 1994. M. Okamoto et al. confirmed the localization of nuclear reactions in the CFP for the first time in 1994. There are several astonishing data sets on the nuclear transmutations (NTs) in very diverse systems from transition-metal hydrides and deuterides such as NiH_x and PdD_x (x \approx 1) to carbon-hydrogen systems

including hydrogen graphite, XLPE and microbial cultures.

To give a unified explanation of the vast and diverse experimental data, we have to follow the orthodox approach to the unknown phenomenon, the phenomenological approach, often used in the history of science. A phenomenological approach using a model based on the experimental facts, the TNCF model, has shown its usefulness to give a unified explanation of the CFP. In the course of the review of typical data sets in this paper, we use the model as a point of view necessary to grasp the total image of the CFP.

Through the review of typical papers on the CFP depicted in this paper, we have given an overview of the science of the nuclear reactions occurring in CF materials (materials where occurs the CFP) demonstrated by vast and dispersed events observed in these 25 years. In short, the science of the CFP is the science of neutrons in CF materials.

1. Introduction

The researches on the CFP have had a long history lasting more than a quarter of a century from the discovery of a part of its vast and diverse contents [Fleischmann 1989, Jones 1989] with experimental data piled up without giving any consistent understanding for them.

To develop the science of the CFP, it is inevitable to have the intimate internal communication of researchers in this field and also communication with scientists in established fields, nuclear physics and solid state physics, closely related to the CFP. The communication with other fields has been given several times in the history, especially in the case of the investigations of the CFP by the committees in the DOE (the Department of Energy), USA as discussed in our recent paper [Kozima 2015 (1)].

In our research field, on the other hand, recent investigation seems to tend not toward the fundamental physics of the CFP but toward the application of this phenomenon with materials having rather complicated components and structures to obtain efficient occurrence of reactions resulting in excess energy and nuclear products. It should be emphasized that the most important direction of the research is, always, scientific and the effective application will follow then the scientific result.

It is meaningful, at present, to review typical papers obtained in this field in rather simple systems in these 25 years and to contemplate the essence of the CFP. We hope this review gives us a condensed material to investigate the fundamentals of the physics of the CFP.

Characteristics of the CFP are pointed out according to following items (1) – (3) in the nuclear reaction (1.1) between a nucleus ${}^{A}_{Z}X$ and another nucleus ${}^{A'}_{Z'}X'$ in the CF materials (materials where occurs the CFP);

 ${}^{A}{}_{Z}X + {}^{A'}{}_{Z'}X' \to {}^{A''}{}_{Z''}X'' + {}^{A'''}{}_{Z'''}X''' + Q, (Z + Z' = Z'' + Z''', A + A' = A'' + A''')$ (1.1)

(1) Probability *P* of the reaction (1.1) in free space depends strongly on the initial mutual energy of the left-hand side particles ε as expressed in the following formula; $P \sim C(\sqrt{\mu/\hbar}) \exp[-\int \sqrt{(V(x) - \varepsilon)} dx],$ (1.2)

where V(x) is the mutual potential energy between the nuclei ${}^{A}_{Z}X$ and ${}^{A'}_{Z'}X'$ and *C* is a constant

- (2) The released energy Q with values of about a few MeV should be participated by nuclei (photons) ${}^{A''}_{Z''}X''$ and ${}^{A'''}_{Z'''}X'''$ in free space.
- (3) Qualitative (or Statistical) reproducibility of the reaction (1.1) is expected if we can arrange the macroscopic initial condition for the reaction.

In reality, the presumptions of the pioneers in this field who published the first paper in 1989 [Fleischmann 1989] were realization of the following reactions (in free space) in deuterated transition metals PdD_x and TiD_x at around the room temperature;

$$d + d \rightarrow {}^{4}_{2}\text{He}^{*} \rightarrow t (1.01 \text{ MeV}) + p (3.12 \text{ MeV}),$$
 (1.3)

$$\to {}^{3}_{2}\text{He} (0.82 \text{ MeV}) + n (2.45 \text{ MeV}), \qquad (1.4)$$

$$\rightarrow {}^{4}_{2}\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}).$$
(1.5)

There are too many obstacles to realize the reactions (1.3) - (1.5) in room temperature solids and the presumptions have been denied by almost all scientists in the world and even the experimental data have been disbelieved as a whole. The obstacles are listed up as follows:

(1a) The value P in Eq. (1.2) estimated for a room-temperature solids PdD_x , for instance, is too small to be measured by ordinary measurements,

(2a) The reaction products with energies of a few MeV order have not observed in commensurate with the reaction formulae (1.3) - (1.5),

(3a) The reproducibility has not been obtained to satisfy ordinary experimental conditions.

To overcome the difficulties (1a) - (3a) pointed out above for the reactions in the CF materials, we have proposed a phenomenological model (the TNCF model) [Kozima 1994] based on the experimental data. In our model, the first (1a) of the three obstacles, reaction probability, had been overcome by participation of the neutral particle – neutron – which is free from the mutual potential V(x). The second (2a), ends of the liberated energy, had been resolved by an assumption that the liberated energy Q is absorbed by the lattice of the CF material. And the third (3a), qualitative reproducibility, had been explained by the mechanism of formation of a specific structure of host nuclei and hydrogen atoms in the CF material described by nonlinear dynamics which

inevitably includes complexity such as self-organization and chaotic behavior of atomic components.

Essential premises of the TNCF model (trapped neutron catalyzed fusion model) were explained in our books [Kozima 1998a, 2006] and papers [Kozima 2014a, 2015 (10)] and is summarized as follows;

- (a) Existence of trapped neutrons with thermal energy in CF materials with a density n_n ,
- (b) The trapped neutrons exert nuclear reactions with displaced and foreign nuclei in the CF materials with the same reaction probability in free space,
- (c) The liberated energy in the above reaction is participated by the lattice of the CF material.

The TNCF model has given a unified qualitative and sometimes semi-quantitative explanations to the whole experimental data sets [Kozima 1998a, 2006, 2014a, 2015 (8)]. Especially, the nuclear transmutations with large changes of proton number Z and nucleon number A described by following formula are explained by participation of neutron drops ${}^{A'}_{Z'}\Delta$ composed of Z' protons and (A'-Z') neutrons;

$${}^{A}_{Z}X + {}^{A}_{Z'}\Delta \to {}^{A}_{Z''}X'^{*}(Z' > 1, (A' - Z') > 1).$$
(1.6)

So, it is a valuable viewpoint to summarize the CFP using the TNCF model even if which is not necessarily a sole viewpoint we have to follow in our research in this field.

The next step necessary to establish the science of the CFP from our point of view is justification of premises assumed in the TCNF model using principles of quantum mechanics established in 20th century which surely applicable to atomic and nuclear processes occurring in the CF materials. Our several trials to this direction have been performed and published already [Kozima 2014a, 2016a].

In this paper, we give a review of typical papers on the CFP obtained in rather simple CF materials from our point of view to establish the science of the CFP and discuss the bases of the TNCF model using recent knowledge developed in nuclear physics and solid state physics..

2. DOE Reports 1989 and 2004

It is valuable to introduce the two Reports of the Department of Energy (DOE) issued in 1989 and 2004 [DOE 1989, 2004] as scientific materials on the CFP given by scientists working in relevant research fields [Kozima 2015 (1)].

The Committees in the Department of Energy had been composed of experts in relevant fields to the CFP and their technical opinions should be esteemed. It should, however, be pointed out limitations imposed on them by their duty different from the researchers in this field. Their duty binds them to confine their sight and also their expertise limits their investigation of the data in of the CFP inside their field preventing extension of their sight.

DOE Report 1989 [DOE 1989]

The characterization of the *DOE Report* presented in 1989 [DOE 1989] was given in our book [Kozima 1998a (pp. 3 - 7)]. We can cite our conclusion on the DOE Report 1989;

"Let us point out mistakes in the DOE report.

Conclusion (1) is based on Conclusions (2) ~ (5), and it has no basis if Conclusions (2) ~ (5) are incorrect. The issue of excess heat and fusion products discussed in Conclusion (2) has significance only when D + D reaction is assumed as the main process. This assumption was adopted by the majority of the scientists at that time, including those who discovered cold fusion.

If there is some other mechanism governing the process, this argument is no longer valid. If you are searching for truth, whether one assumption made by a scientist is correct or not has no importance. You should search for the truth based on the fact that the phenomenon did occur. From this point of view, we will show, in Chapters 11 and 12, that it is possible to explain the results of cold fusion experiments without any inconsistency.

Conclusion (3) was based on the fact that the cold fusion phenomenon presented poor reproducibility. However, the reproducibility of a phenomenon is determined by the condition of the entire system, in which the process takes place. Simple analogy from other physical phenomena should not have been used to draw a conclusion. We will also show the reasons for the poor reproducibility and the way to improve it in Chapters 11 and 12.

Conclusion (4) only shows that the interpretations of the discoverers of cold fusion were not appropriate, and it has nothing to do with the truth. It is hard to believe that board members have made such an elementary mistake. It was found later that inside solid, such as Pd or Ti, with a combination of various factors, complex phenomena can occur. There is always such possibility in science. Today, it is quite obvious to everybody. The board members might have forgotten for some reason that natural science is build upon the fact.

Conclusion (5) is similar to Conclusion (4). If any new findings had been denied only because they were contradiction with the existing knowledge, there would have been no progress in science and there will not be any progress in the future.

The discussions expressed in the DOE Report remind us Procrustes' bed. As

Procrustes used his bed as an absolute standard to measure heights of his captives, the critiques against the cold fusion used d - d reaction as an inevitable standard to judge anomalous events." [Kozima 1998a]

A scientific spirit in the *Report* is sparkling in the following sentence added as a comment (believed to be written by N.F. Ramsey);

"- - - as a result, it is difficult convincingly to resolve all cold fusion claims since, for example, any good experiment that fails to find cold fusion can be discounted as merely not working for unknown reasons. Likewise the failure of a theory to account for cold fusion can be discounted on the grounds that the correct explanation and theory has not been provided. Consequently, with the many contradictory existing claims it is not possible at this time to state categorically that all the claims for cold fusion have been convincingly either proved or disproved - - - ." [DOE 1989]

DOE Report 2004 [DOE 2004]

The DOE Report 2004 [DOE 2004] has a different character from that of 1989. The new *Report* was issued according to the request presented by several CF researchers as a document [Hagelstein 2004].

"The Department of Energy's (DOE) Office of Science (SC) was approached in late 2003 by a group of scientists who requested that the Department revisit the question of scientific evidence for low energy nuclear reactions. In 1989 Pons and Fleischman first reported the production of "excess" heat in a Pd electrochemical cell, and postulated that this was due to D-D fusion (D=deuterium), sometimes referred to as 'cold fusion.' The work was reviewed in 1989 by the Energy Research Advisory Board (ERAB) of the DOE. ERAB did not recommend the establishment of special programs within DOE devoted to the science of low energy fusion, but supported funding of peer-reviewed experiments for further investigations. Since 1989, research programs in cold fusion have been supported by various universities, private industry, and government agencies in several countries." [DOE 2004]

"Mail Review Charge Letter of DOE" says;

"Enclosed is the summary document and appendix material related to the review of recent scientific reports of low energy nuclear reactions (LENR) in metal matrices,* currently being conducted for the Office of Science by the Offices of Basic Energy Sciences and Nuclear Physics in the Department of Energy on the recent scientific reports of Low Energy Nuclear Reactions (LENR). The goal of the review will be to generate a report on the status of the research field for the Director and the Principal Deputy Director of the Office of Science. The report will be written by DOE federal staff based on the individual inputs from members of a DOE empaneled review team." [DOE 2004]

*The scientific reports are the one, the title, authors and abstract of which are cited as follows;

"New Physical Effects in Metal Deuterides

P.L. Hagelstein, M.C.H. McKubre, D.J. Nagel, T.A. Chubb, and R.J. Hekman, Abstract

The experimental evidence for anomalies in metal deuterides, including excess heat and nuclear emissions, suggests the existence of new physical effects." [Hagelstein 2004]

According to the limited evidences given to the DOE as clearly written in the above *Abstract*, the material is confined to the "*The experimental evidence for anomalies in metal deuterides*" and does not include the data obtained in the protium systems. Therefore, the material given to the DOE is necessarily an incomplete one to show the cold fusion phenomenon as a whole. However, the Report [DOE 2004] had merit to evaluate positive phases of the CF researches after the DOE Report 1989 [DOE 1989].

Conclusion of DOE is cited as follows;

"While significant progress has been made in the sophistication of calorimeters since the review of this subject in 1989, the conclusions reached by the reviewers today are similar to those found in the 1989 review.

The current reviewers identified a number of basic science research areas that could be helpful in resolving some of the controversies in the field, two of which were: 1) material science aspects of deuterated metals using modern characterization techniques, and 2) the study of particles reportedly emitted from deuterated foils using state-of-the-art apparatus and methods. The reviewers believed that this field would benefit from the peer-review processes associated with proposal submission to agencies and paper submission to archival journals." [DOE 2004]

It should be cited one of the positive comments in the Report as follows;

"It is now clear that loading level and current density thresholds are required in order to observe excess heat in these experiments. The values are consistent regardless of the approach used and the laboratory where the experiment was conducted. Early failures to reproduce the heat effect were, in part, due to not meeting these requirements. It has also been found that thermal and current density transients, which are thought to effect the chemical environment such as deuterium flux, can trigger heat "events". SRI has published an expression for the correlation between excess power and current density, loading, and deuterium flux. These discoveries have led to a better understanding of the phenomena and more reproducibility." (Reviewer #9)

One of the important results of the CFP not taken up in the DOE Report 2004 is the nuclear transmutations in protium and deuterium systems. The nuclear transmutation (NT) is an astonishing event suggesting a new state of matter in the CF materials (materials responsible to the CFP) entirely different from the states of matter we have had known in physics and chemistry developed in the 20th century.

The data of nuclear transmutations in the CFP are summarized in following books and papers [Kozima 1998a (Chapter 9), 2006 (Section 2.5), 2014a, Storms 2007 (Section 4.5)] and the stability effect found in the data of nuclear transmutation is explained in following papers and books [Kozima 2005, 2006 (Section 2.11), 2012a].

3. Typical Papers in the History of the Cold Fusion Phenomenon (CFP)

In the history of science, there were several papers which had given decisive impacts on the development of a science; an example of these papers is the paper "*Zur Elektrodynamik bewegter Körper*", *Annalen der Physik* **17**: 891 (1905)" by A. Einstein which had given Copernican revolution on the physical view of energy and matter.

In the field of the CF research (the research on the CFP), we could not find out such epoch making papers, or papers inducing a paradigm revolution of material science in 20th century physics, comparable to that by A. Einstein. However, we have several typical papers showing evidences of nuclear reactions in CF materials composed of specific elements and hydrogen isotopes. It is inevitable to take up these papers as a foundation for development of new science for the CFP. In this section, we introduce these typical papers and also some succeeding papers which confirmed the essence of the typical papers.

3.1 Detection of ⁴₂He by Morrey et al.

One of the crucial evidences of nuclear reactions in the CFP is detection of ${}^{4}_{2}$ He accompanied to excess energy according to the reaction (1.5) if there occur d - d fusion reactions in CF materials. The trial to check the nature of the apparent nuclear reactions in the CFP had been performed as early as 1989 just after the pioneering paper by

Fleischmann et al. The Pacific Northwest Laboratory (PNL) presided the six laboratories chosen by the University of Utah (U-o-U) to check existence of ${}^{3}_{2}$ He and ${}^{4}_{2}$ He in Pd samples provided by the U-o-U. Their experimental result was published in the *Fusion Technology* (ISSN:0748-1896) published by the American Nuclear Society [Morrey 1990]. For the benefit of readers, this paper is posted at the CFRL website next to the CFRL News No. 87:

http://www.geocities.jp/hjrfq930/News/news.html

They measured no ${}^{3}_{2}$ He and a scanty ${}^{4}_{2}$ He in the surface region of a width about 25 μ m with an amount incommensurate to the reported excess energy from the sample according to the presupposed d - d nuclear fusion reactions (1.3) – (1.5) [Fleischmann 1989].

Their conclusion is summarized as follows:

"It cannot be proven that the minimal excess heating in one of the rods reported by Fleischmann and Pons can be attributed to the formation of ⁴He, although the possibility that some ⁴He could have formed during electrolysis cannot be ruled out. If ⁴He were generated, the mechanism must be surface related, not bulk related. No attempt was made to measure any helium or tritium hat might have left the cathode surface as gas during electrolysis. The results presented cannot, unfortunately, confirm the existence or nonexistence of cold fusion via helium production. However, they provide a basis for follow-on experiments that should lead to a final conclusion." [Morrey 1990]

This conclusion might be accepted, in general, to show the negative evidence against the CFP induced by the mechanism the d - d fusion reaction. However, it is absurd to deny experimental results that are in contradiction to the presupposed conclusion and to try repeatedly to find a result in accord to it. In science, we have to rely on the confirmed facts irrespective of the supposed anticipation.

When we accept the experimental results frankly and investigate them without prepossession of the reactions (1.3) - (1.5), we can construct a model consistent with many experimental data including the one by Morrey et al. [Kozima 1997, 1998a, 1999, 2006, 2014b].

We have to notice here the difficulty in high-precision determination of helium in samples. W.B. Clarke was a specialist in measurement of a trace of helium, for instance the blood helium concentration. He was asked to measure the helium content in a sample supplied by M.C.R. McKubre et al. of SRI (Stanford Research Institute). The result was not consistent with the excess energy result obtained in the same sample if

we assume the reactions (1.3) - (1.5). However, our analysis of the data had given a consistent explanation of the data sets by Clarke et al. and by McKubre et al. as presented at ICCF9 [Kozima 2003].

The short survey of the history of ${}^{4}_{2}$ He detection given above clearly shows us a simple fact that researches in science should rely only on experimental data leaving our presumptions. This proper common sense seems to be a rather weak current in the CF research field, unfortunately. We have to be scientific above all else.

Another remarkable result obtained by Morrey et al. is the surface nature of the CFP. "If ⁴He were generated, the mechanism must be surface related, not bulk related." [Morrey 1990]. This characteristic of the nuclear reactions in the CFP has been confirmed by many experiments afterwards and explained by our model (or has been used to construct our model) [Kozima 1999].

It should be given a comment on the claim that the measurements of helium and excess energy have confirmed occurrence of the reaction (1.5) in room-temperature solids as insisted several times (e.g. [Hagelstein 2004]):

 $d + d \rightarrow {}^{4}_{2}\text{He}^{*} \rightarrow {}^{4}_{2}\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}).$ (1.5) They say that the excess energy Q corresponds to the amount of ${}^{4}_{2}\text{He}$ predicted by the reaction (1.5). One of such claims was the paper [Hagelstein 2004] presented to DOE and cited in the DOE Report 2004 [DOE 2004].

Difficulty of occurrence of the reaction (1.5) in solids giving the liberated energy to the lattice of the solids was discussed in the DOE Report 2004 [DOE 2004]. They rejected acceptance of the claim from theoretical and experimental bases.

We would like to point out a difficulty of the claim from experimental point of view based on the consistency of experiments. As we have discussed the diversity of experimental data in the CFP, there are very many events in the CFP resulting in various nuclear transmutations accompanying corresponding excess energy. In these events, we can find out cases where were measured helium-4 ${}^{4}_{2}$ He, tritium ${}^{3}_{1}$ H, and excess energy *Q* simultaneously [Chien 1992, Gozzi 1995] in addition to other results detecting these and other observables individually. Then, the above claim means that there is only the reaction (1.5) in the PdD_x systems excluding any other reactions. This claim is inconsistent with the experimental facts that there are huge data sets showing occurrence of nuclear reactions other than the reaction (1.5). It is difficult to consider exclusive occurrence of the reaction (1.5) in a specific system engaged by the authors of the paper insisting the claim.

3.2 Nuclear Transmutations in Protium Systems and Observation by

Bush and Eagleton

The first report of the CFP in a protium system is, as far as I know, the one by R.L. Mills and S.P. Kneizys appeared in the *Fusion Technology* in 1991 [Mills 1991]. The second will be that by R.T. Bush also published in *Fusion Technology* in 1992 [Bush 1992]. These papers reported the excess heat generation in protium systems. It is interesting to notice their interpretation of their results to explain the unexpected nuclear reactions (resulting in the observed enormous amount of the excess energy). The parallelism of the d + d fusions supposed by Fleischmann et al. in the deuterium system should be the p + p reactions in the protium system, which were too far from their common sense in physics and they assumed specific mechanisms each other. Mills et al. assumed a specific mini-hydrogen (hydrogen atoms with fractional quantum numbers) and R.T. Bush assumed a direct fusion reaction of a proton and an alkaline nucleus (alkali-hydrogen fusion).

We have classified experimental data into two classes, direct and indirect evidences of nuclear reactions resulting in the CFP, to make clear the importance of the experimental data [Kozima 1998a, 2006]. The excess energy (heat) is classified in the indirect evidence of the nuclear reactions if its amount is too much to explain by known physical or chemical processes without nuclear reactions. Therefore, it was desirable to observe any direct evidence in protium systems to declare the existence of the CFP in them.

The first direct evidence of the nuclear reactions in protium systems was measured by R.T. Bush and R.D. Eagleton in 1993 [Bush 1993, 1994]. For the benefit of readers, we have uploaded the paper [Bush 1994] in the CFRL website next to the CFRL News. No. 89;

http://www.geocities.jp/hjrfq930/News/news.html

We have analyzed their data using our TNCF model applicable to deuterium and also protium systems and have given a semi-quantitative explanation of their observation of transmuted nuclei [Kozima 1995, 1996a, 1998a (Sec. 9.1b), 2010b, 2015(3), Ohta 1996].

It is important to know a history of the journalism around the CF research. It is well known that the *Fusion Technology*, the authorized international journal of the American Nuclear Society, played a very important role in the promotion of CF researches. The editor of the journal at that time was Prof. G.H. Miley and his judgment made possible to publish many papers on the CFP. The editor of the journal changed from Prof. Miley to another in the year of 2001 and the name of the journal changed to *Fusion Science and Technology*. After this change, it seems the character of the journal has become less

scientific. A Comment by G.H. Miley at his retirement from the editor is posted at CFRL website: <u>http://www.geocities.jp/hjrfq930/FTEssay/Essays/Miley.htm</u>

Researches of the CFP in protium systems have been accelerated by the papers by R.L. Mills and R.T. Bush and many papers have presented at international conferences and published in international journals. In ICCF3 held in Nagoya, Japan in 1993, there are following pioneering papers in this genre by Notoya and Enyo [Notoya 1993], Ohmori and Enyo [Ohmori 1993] and Srinivasan et al. [Srinivasan 1993]. By the way, it should be noticed a short essay "Open Minded Attitudes to the Science" by Michio Enyo, one of the authors cited above, which is posted at a following page of the CFRL website:

http://www.geocities.jp/hjrfq930/FTEssay/Essays/Enyo.htm

Now, it is not necessary to mention the reality of the CFP in protium systems when there are too many experimental data sets showing nuclear reactions in them [Kozima 1998a, 2006, Storms 2007]. The most elaborate works on Ni-H systems have been performed by Focardi et al. in Italy [Focardi 1994, Campari 2000, 2006] and Enyo et al. in Japan [Ohmori 1993, 1996, 1997]. These data on protium systems had been successfully analyzed by our model as presented in books and papers [Kozima 1998a, 2006, 2010b].

From our present view, it is interesting to look for a first person or people who observed the CFP in protium systems. In relation to this question, we find an episode told by opponents against the CFP. The episode is given in Appendix which tells us the observation of excess heat in protium system by S. Pons for the first time in the history.

3.3 Energy Spectrum of Neutrons in the CFP and Observation by Jones et al.

It is well known that the first observation of the energy spectrum of neutrons emitted from cold fusion materials (CF materials) was performed by Jones et al. [Jones 1989] in BYU in the State of Utah, USA. A unified explanation of their data in addition to the data of excess energy observed by Fleischmann et al. was given by us [Kozima 1997, 1998a].

Even if they insisted the discovery of the $E_n = 2.45$ MeV neutrons emitted by the reaction Eq. (1.4) of *d*-*d* fusion reactions, there remained possibility of higher energy neutrons at channels 230 – 300 ($E_n = 5.8 - 7.5$ MeV) in their data cited below as Fig. 3.3.1.



Fig. 3.3.1 Neutron energy spectrum observed by Jones et al. [Jones 1989]. The channel 100 corresponds to the neutron energy 2.45 MeV.

In reality, Takahashi et al. [Takahashi 1991] and Bressani et al. [Bressani 1991] confirmed clearly the existence of higher energy neutrons up to 10 MeV as shown in Figs. 3.3.2 and 3.3.3. This is an evidence of other nuclear reactions than d-d fusion reactions Eq. (1.3) – (1.4) occurring in the CF materials.

Details of this meaning have been discussed in our books and papers published by now [Kozima 1998a (Sec. 11.4), 1998b, 2006 (Sec. 3.3.6), 2015 (4)].



Fig. 3.3.2 Energy spectrum observed by Takahashi et al. [Takahashi 1991].



Fig. 3.3.3 Energy spectrum observed by Bressani et al. [Bressani 1991]

3.4 Measurements of Excess Energy and the Works by McKubre et al.

The measurement of excess energy, one of the indirect evidences of the CFP, was the most reliable result among others in the content of the pioneering paper by Fleischmann et al. published in 1989 [Fleishmann 1989, Kozima 1998a, 2006]. They used PdD_x, one of the typical cold fusion materials (CF materials) of the deuterium system. (By the way, we would like to recall that another CF material used frequently in the CFP research is NiH_x as discussed in our paper [Kozima 1998a, 2006].

The experimental data sets published by McKubre et al. in 1993 and 1994 in the same PdD_x system as that by Fleischmann et al. with a more sophisticated apparatus are the most extensive one ever obtained [McKubre 1993, 1994a, 1994b]. We have investigated the elaborate result by McKubre et al. and elucidated its indispensable value for the science of the CFP [Kozima 2015 (5)].

One of the figures in the paper by McKubre et al. [McKubre 1993] reproduced in Fig. 3.4.1 shows the elaborateness of their work. This point had been evaluated already in DOE Report [DOE 2004] as cited in Section 2:

"While significant progress has been made in the sophistication of calorimeters since the review of this subject in 1989, - - - ." [DOE 2004]



Fig. 3.4.1 Variation of excess power with loading ratio D/Pd observed by McKubre et al. [McKubre 1993].

The extensive experiments by McKubre et al. [McKubre 1993, 1994a, 1994b] have revealed various important features of the CFP from the dependence of the average excess power P on the cell current i and on the loading ratio x to complex behavior (complexity) of the events of the excess power generation. The latter behavior should be taken as a fundamental but not accidental or artifactual feature of the CFP which resolves the problem of absence of the quantitative reproducibility (or rather existence of the qualitative reproducibility) of the experimental results as discussed in Section 3.8 below in addition to our works published before [Kozima 2006, 201a].

3.5 Determination of Localized Nuclear Reactions and the Experiment by Okamoto et al.



Fig. 3.5.1 Examples of depth profiles for each element [Okamoto 1994]

The excellent analysis of Pd cathode provided by Fleischmann et al. to detect ${}^{4}_{2}$ He in the surface layer of a width of about 40 µm by Morrey et al. had been discussed already in our paper [Kozima 1997] and our CFRL News No. 88; http://www.geocities.jp/hjrfq930/News/news.html.

It is possible to say that this data obtained by Morrey et al. was the first determination of the local nature of the CFP from our present knowledge as we had discussed there. We have explained their experimental result by our TNCF model where ${}^{4}{}_{2}$ He was generated by Li-*n* reactions in the surface layer PdLi_x [Kozima 1996b, 1997, 1998 (Sec. 11.8a), 1999, 2011] as suggested by the experimental data obtained by Okamoto et al. [Okamoto 1994].

The data given in Fig. 3.5.1 [Okamoto 1994] shows the nuclear transmutation from ${}^{27}{}_{13}$ Al to ${}^{28}{}_{14}$ Si in the surface layer on the Pd cathode in Pd/D₂O+LiOD system. This data was semi-quantitatively explained by the TNCF model [Kozima 1998a (Sec. 11.11e)].

There have been many investigations on the localization of nuclear transmutations in the CFP (e.g. [Bockris 1995]) which are summarized in our recent papers [Kozima 2011, 2015 (6)]. Most remarkable one of the many data sets showing the localization of nuclear reactions is the data by Iwamura et al. presented at ICCF12 [Iwamura 2006]. The data given in Fig. 3.5.2 [Iwamura 2006] shows localized generation of Pr from Cs in their specific structure called "Pd complex." Details of analysis of their data including that one shown in Fig. 3.5.2 are given in our paper [Kozima 2011].



Fig. 3.5.2 Nuclear transmutations from Cs to Pr [Iwamura 2006].

3.6 Detection of Tritium in CF-Materials by Packham et al.

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 1989] in which many experts on the relevant fields of science to the CFP contributed to investigate extensively scientific value of "*the apparent observations of cold fusion and significant quantities of energy from these phenomena*" since April to November, 1989.

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 Report [DOE 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 [Kozima 2015 (7)].

The tritium measurements have been performed using several types of CF materials;

(1) Pd/D/Li and PdSi_x/D/Na electrolytic systems. The most popular one is Pd metals hydrogenated by electrolysis with electrolyte $D_2O + LiOD$. 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_x/D/Na system was used by Iyengar et al. [Iyengar 1989].

(2) Pd/D_2 system with Pd powder. Iyengar et al. [Iyengar 1989] and Claytor et al. [Claytor 1993, Tuggle 1994].

(1) Pd/D/Li and PdSi_x/D/Na Electrolytic Systems.

Many measurements of tritium have been performed with Pd/D/Li system and one by Iyengar et al. with PdSi_x/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 D₂O + 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 5×10^{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_x/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. \times 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 (Pd, PdSix, Ti samples with various shapes which occluded deuterium) 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 individually, 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 1989a (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/D₂ and Pd/D₂ Gas Loading Systems.

In the experiment by Iyengar et al., Pd samples were either in the form of Pd-black powder or Johnson & Matthey Pd-Ag foils [Iyengar 1989 (Sec. 7.1)]. In the case of Ti/D₂ system, they observed *n* and *t*. The quantity of D₂ absorbed in Ti could be measured from the observed pressure drop. This corresponded to $\approx 10^{19}$ molecules of D₂ gas, indicating a gross (D/Ti) ratio of hardly 0.001. However, it is believed that most of the absorbed D₂ 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/D₂ system [Tuggle 1994], Pd samples were powder (small (0.3 to 0.5 μ m) spheres that form chains or agglomerates up to 30 μ m in dia.), foil (220 micron thick) and wire. Four types of cells have been made: (a) those with palladium powder and silicon powder, (b) those with palladium foil and silicon powder, (c) those with palladium foil and silicon wafers, and (d) one 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. 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 or plasma." [Tuggle 1994]

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 and papers [Kozima 1998a (Sec. 11.7), 2004a, 2006 (Secs. 2.6 and 3.3), 2014b, 2015 (7)]. In the "Sec. 6.4 Tritium" of the Discovery of the Cold Fusion Phenomenon [Kozima 1998a], we introduced the works by Srinivasan et al., Storms and Talcot, Tuggle et al. [Tuggle 1994], 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 + phonons (6.25 MeV),	(3.6.1)
$n + {}^{6}_{3}\text{Li} = {}^{4}_{2}\text{He} (2.1 \text{ MeV}) + t (2.7 \text{ MeV}),$	(3.6.2)

where the phonons in the Eq. (3.6.1) are supposed to be shared by the CF material as explained in Introduction as a premise (2a). This process is supposed to be realized through neutrons in the neutron band coupled to the lattice by the super-nuclear interaction [Kozima 2006 (Sec. 3.7), 2014b, 2016a].

Other data sets on the tritium detection have been discussed in our paper [Kozima 2015(7)]

3.7 Nuclear Transmutations in Carbon-Hydrogen Systems – Biotransmutation, Hydrogen Graphite, and XLPE (Cross-Linked Polyethylene) –

It was an astonishing fact to find many new elements in a system where carbon arc was discharged between graphite electrodes in water [Hanawa 2000]. The astonishment has been extended further in other carbon-hydrogen systems by detections of nuclear transmutations in biological systems [Vysotskii 2009a] and in cross-linked polyethylenes [Kumazawa 2005]. In this section, we give a brief explanation of nuclear

transmutations in these carbon-hydrogen systems according to our recent paper [Kozima 2015(8)].

3.7.1 Nuclear Transmutation in Hydrogen Graphite HC_x (x = 6 - 8)

In the carbon arc system with graphite electrodes in water, there are found many kinds of elements in addition to the most abundant iron (Fe); Si, S, Cl, K, Ca, Ti, Cr, Mn, Co, Ni, Cu, Zn, and possibly heavier elements

In the experiments performed by T. Hanawa, he found various new elements and changes of quantities of elements in the product of carbon arc with graphite electrodes in light water using energy dispersive X-ray spectrometry, method of X-ray fluorescence (XRF) and particle induced X-ray emission [Hanawa 2000].

The result revealed increases of elements in the system and appearance of many kinds of new elements; Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Co, Cu, Zn, and possibly heavier elements. The relative abundance including null, however, varied case by case. Furthermore, the dominant product Fe was found only in the larger debris.

Among XRF inspections applied to arc traces of used electrodes, an anode showed metallic elements, which suggest that transmutation reactions took place on the anode surface.

The experimental results obtained in carbon arc are very difficult to understand similarly to the data obtained in XLPE and biological systems introduced following subsections. However, we may be able to treat the data in these carbon-hydrogen systems using our TNCF model as explained in this section.

To understand the experimental data obtained in carbon arc, we have to know the structure of the graphite electrodes used in the arcing in water. Structure of graphite is shown in Fig. 3.7.1.



Fig. 3.7.1 Side view of layer stacking of graphite (after Wikipedia).

It is well known that graphite readily oxidizes in atmospheres containing oxygen to form CO_2 at temperatures of 700 °C and above. Therefore, we may suppose that the electrodes of arc are covered with CO_2 layers on the surface. Furthermore, H atoms will

be absorbed into the volume to form an intercalation compound, hydrogen graphite HC_x , (x = 6 - 8 ?) which is supposed to have similar structure to that of potassium graphite KC_8 or calcium graphite CaC_6 , which is shown in Fig. 3.7.2.



Fig. 3.7.2 Structure of CaC₆ (after Wikipedia): violet spheres represent Ca nuclei between layers of carbon nuclei (grey spheres). We may imagine the structure of hydrogen graphite HC_x (x = 6 - 8 ?) which is not determined yet referring to this structure of CaC₆.

We may have then a superlattice made of a carbon sublattice of graphite and a hydrogen sublattice occluded between carbon layers in the graphite electrodes covered by CO_2 layer. The HC_x superlattice may have a structure similar to the superlattice CaC_6 shown in Fig. 3.7.2. If the hydrogen graphite HC_x forms such a superlattice considered above, there are formed the CF-matter which participate the CFP by the mechanism proposed in our books [Kozima 2004b, 2006 (Sec. 3.7)] and papers [Kozima 2005, 2016a]. Thus, the product elements observed in the system of carbon arc in water are explained by our TNCF model as a result of the nuclear transmutation catalyzed by the trapped neutrons.

3.7.2 Nuclear Transmutation in XLPE

The excellent experimental data on the nuclear transmutation in cross-linked polyethylene had been obtained by Kumazawa et al. for more than 10 years from 2005 [Kumazawa 2005, 2006, 2012].

To show the essential feature of the atomic alignment in XLPE, we show its molecular structure in Fig. 3.7.3:



Fig. 3.7.3 Lattice structure of XLPE orthorhombic lattice with lattice constants, a = 7.40 Å (740 pm), b = 4.93 Å (493 pm), c = 2.53 Å (253 pm) [Kozima 2010c (Fig. 5)].

The experimental data obtained by Kumazawa et al. [Kumazawa 2005, 2006] have successfully been analyzed by the TNCF model as explained by our papers [Kozima 2010c, 2016b]. The superlattice of C and H in the XLPE works to generate neutron bands according to the mechanism figured out in the TNCF model and the neutrons in the bands thus formed induce nuclear reactions and produce new elements observed by Kumazawa et al. Recent data of gamma emission from XLPE [Kumazawa 2012] is analyzed using the TNCF model successfully and the result is presented at this Conference [Kozima 2016b].

3.7.3 Nuclear Transmutations in Biological Systems (Biotransmutations)

The experimental data sets on the biotransmutation have been obtained in these about 20 years mainly by V.I. Vysotskii and his collaborators [Vysotskii 1996, 2009a, 2009b, 2015]. There are data sets showing (1) production of ${}^{57}_{26}$ Fe from ${}^{55}_{25}$ Mn [Vysotskii 1996, 2015] and also (2) acceleration of the decay of radioactive nuclei ${}^{157}_{55}$ Cs, ${}^{140}_{56}$ Ba and ${}^{140}_{57}$ La in several microbial cultures [Vysotskii 2009b, 2015]. For the benefit of readers, the paper [Vysotskii 2015] is posted at the CFRL website next to the CFRL News No. 94:

http://www.geocities.jp/hjrfq930/News/news.htlm

Experiments were conducted using several bacterial cultures (*Bacillus subtilis GSY 228, Escherichia coli K-1, Deinococcus radiodurans M-1*) as well as the yeast culture *Saccharomyces cerevisiae T-8*. Selection of these cultures was motivated either by their experimentally proven ability to grow in the heavy water based media or by the prospect

of using the radiation-stable culture *Deinococcus radiodurans M-1* in transmutation processes given the presence of powerful radioactive fields, as was noted earlier [Vysotskii 2009a].

To show the general idea of the molecular structure of bacteria, we show first the cell structure of a gram positive bacterium in Fig. 3.7.4 and the structure of peptidoglycan, a polymer consisting of sugars and amino acids that forms a mesh-like layer outside the plasma membrane of most bacteria, forming the cell wall, is shown in Fig. 3.7.5.



Fig. 3.7.4 Cell structure of a gram positive bacterium (after Wikipedia).

Peptidoglycan is made up of a polysaccharide backbone consisting of alternating N-Acetylmuramic acid (NAM) and N-acetylglucosamine (NAG) residues in equal amounts.



Fig. 3.7.5 The structure of peptidoglycan (after Wikipedia).

The complex structures of biological cells, a part of which are shown in Figs. 3.7.4 and 3.7.5, have regular arrays of molecules made of carbon and hydrogen, similar to the

array found in XLPE in rather simple form discussed in the previous section. The superlattices found in these biological systems might be able to generate neutron bands as in the case of XLPE where we can assume the CF matter to induce the CFP as observed by Vysotskii et al. Details of the treatment are given in another paper presented at this Conference [Kozima 2016c].

3.8 Qualitative Reproducibility and Complexity in the CFP

The reproducibility of events in the CFP has been one of serious controversies between pros and cons of the CFP. Even in CF researchers, there are many who consider the events in the CFP should be reproducible as in the simple systems described by linear differential equations. However, the experimental data obtained by now have shown that there is no quantitative reproducibility such as observed in simple systems.

On the other hand, it is possible to say that the events in the CFP are qualitatively reproducible, or statistically reproducible, as we know well engaging in investigation of the CFP. Therefore, we have to find out the reason why the events in the CFP are not quantitatively reproducible but are qualitatively reproducible. Our approach to this problem has been given in papers and books [Kozima 2006 (Sec. 3.8), 2010a, 2012a, 2012b, 2014b, 2015 (9)] assuming existence of complexity in the CF materials which inevitably results in qualitative reproducibility or irreproducibility in the worst case.

3.8.1 CF material and CF-matter

The field where the CFP occurs may be a special one because there occur events incredibly different from those occurring in other fields of established science e.g. solid state physics, we have to use specific terminology which does not have a civil right in other branches of science. We define "the *cf-matter*" as the necessary condition (or state) for occurrence of the CFP in a "*CF material*" or "*CF substance*" (a solid material composed of a host element (e.g. C, Ti, Ni, Pd, etc.) and a hydrogen isotope (H or/and D) where occurs the CFP).

3.8.2 Construction and Destruction of the CF-matter

Construction and destruction of the "CF-matter" (a state made of neutrons in neutron bands and protons) occur according to the atomic processes (microscopic processes) in a CF material arranged by an experimental setup (macroscopic processes) in a dynamical, non-equilibrium system composed of multi-component inhomogeneous materials.

The construction is governed by essentially stochastic (or statistical) atomic

processes occurring in inhomogeneous materials composed of a solid (transition metals or carbon) and hydrogen atoms H (and/or deuterium atoms D). The atomic processes include adsorption of H (D) on the surface of solids, absorption into and occlusion in the solids of H (D), formation of an intermetallic compound (e.g. PdD, NiH, etc.), or formation of a regular array of a hydrocarbon (e.g. XLPE, microbial cultures, etc.) where exist stochastic processes (diffusion) and/or self-organization of a stoichiometric compound in local area from non-stoichiometric solution.

The macroscopic arrangement of an experimental initial condition does not completely determine the microscopic initial condition at all and there is a vast freedom not determined by the arrangement which results in variety of CF materials. The variety itself may produce different effects in the sample at and after nuclear reactions between components of the CF material, CF-matter and displaced or foreign atoms.

Furthermore, the self-organization is not controlled by the macroscopic initial condition at all and therefore the resulting CF-matter is not controllable from outside.

3.8.3 Unpredictability and Irreproducibility

There have been a long history of unresolved disputes between pros and cons about the reality of the CFP since the first stage of the investigation when the paper by Fleischmann et al. [Fleischmann 1989] and the *DOE Report 1989* [DOE 1989] were published in 1989. However, it seems that there is a misunderstanding of the meaning "irreproducibility" in science which will be resolved by consideration of the relation of cause and effect in proper concepts.

It will be possible to say that the concept "unpredictability" in theoretical context corresponds to the "irreproducibility" in experimental situation. We say the effect is unpredictable when we cannot predict the result (effect) for a definite initial condition (cause) for a system. In this case, a cause does not give a definite effect. We say the effect is irreproducible when we cannot obtain the same result (effect) for a (supposedly) the same experimental condition for a system.

The cause-effect correspondence (relation) for a physical process is divided into three cases: Effect with (1) "one-to-one" correspondence between them, (2) "one-to-several" correspondence with a probability, and (3) "one-to-none" (or "one-to-some" effects) correspondence with by chance (or without any definite probability).

These cases are expressed by the predictability with (1) a quantitative probability with a definite value, (2) a qualitative probability with statistical values, and (3) zero probability for the effect.

Correspondingly, the three cases are expressed experimentally by (1) a quantitative reproducibility, (2) a qualitative reproducibility, and (3) irreproducibility.

Here, in the CFP, are two causes of unpredictability (and therefore irreproducibility). The first is the stochastic processes in the formation of CF materials and the second is the self-organization of cf-matter in the CF materials including enough amount of hydrogen isotopes in solids.

Destruction of the cf-matter is induced by the CFP itself that makes the components of the CF material shift from the optimal ones for the CFP and also destroys the structure of the CF material by heat and dynamical impact by particles produced by nuclear reactions. The destruction of the CF-matter is another cause of irreproducibility and unpredictability.

3.8.3.1 Qualitative Reproducibility or Statistical Reproducibility

Unpredictability in theoretical context means irreproducibility in experimental context. We use these words interchangeably in the following discussions. In the following subsections, we discuss fundamental processes in the CFP which may influence on the reproducibility of events which occur in complex CF materials.

3.8.3.2 Macroscopic States and Microscopic States

It is impossible to control microscopic states by defining macroscopic states in principle. Furthermore, it is impossible to determine exact states by an experiment without any error. This situation is described clearly in relation to the unpredictability due to instability or chaotic nature of the system in the texts on the nonlinear dynamics.

In the linear dynamical systems where we have mainly treated classical physics, we can say a following expression for the cause-effect relation:

"Measurements could never be perfect. Scientists marching under Newton's banner actually waved another flag that said something like this: Given an approximate knowledge of a system's initial conditions and an understanding of natural law, one can calculate the approximate behavior of the system." [Gleick 1987 (p. 14-15)]

Many researchers in the CFP belong to these who "said something like this."

The expression has to be altered by a following sentence when there is nonlinearity in the system;

"The often repeated statement, that given the initial conditions we know what a deterministic system will do far into the future, is false. Poincaré (1892) knew it was false, and we know it is false, in the following sense: given infinitesimally different starting points, we often end up with wildly different outcomes. Even with the simplest

conceivable equations of motion, almost any non-linear system will exhibit chaotic behaviour. A familiar example is turbulence." [Cvitanovic 1989 (p. 3)]

The problem in the predictability is expressed in the above sentence "*one can calculate the approximate behavior of the system*." in the deterministic system which presupposed negative Lyapunov exponent [Strogatz 1994 (Sec. 10.5)]. It is shown that there are systems in which this is not true as explained there [Strogatz 1994 (Sec. 10.5)].

3.8.3.3 Averaging of Measured Results on an Effect Observed by an Experiment

Effects are sometimes summed up to make them measurable by macroscopic apparatus which is handled macroscopically (e.g. pressure gauge for gas pressure – induced by extravagant number of molecular impacts on a wall).

For events where we satisfy with their effects averaged over time and space, we do not care about their exact initial condition (which is impossible to get by our limited imperfect ability of measurement) but approximate one which results in approximate behavior and in the same average effect irrespective of their initial condition. Irrespective of minor differences in the initial condition, we can reproduce the almost the same result by averaging over approximate results – attaining reproducibility.

However, there are other cases where we meet an individual event but not the averaged one.

The alpha-decay of Radium-226 ($^{226}_{88}$ Ra) is statistical and its average behavior is described by an equation

 $N(t) = N(0) \exp(-t/\tau), \tag{3.8.1}$

where N(0) and N(t) are the numbers of the nuclei at time 0 and t, respectively. The real decay process is not described by the relation (3.8.1) but stochastic; the signals of a Geiger counter amplifying the discharge caused by alpha-particles reflect the decay process of the ${}^{226}_{88}$ Ra placed near the counter.

In this case, the signal of a Geiger counter is not described by a differential equation (3.8.1) but by a difference equation. Suppose that each signal of the Geiger counter gives a tremendous amount of water that we have to treat as fast as possible, we cannot wait several signals to be averaged over them. Then, the averaging of the signals is nonsense and the individual event is meaningful. The situation we met in the CFP may correspond to the latter example described above. And averaging and therefore reproducibility has nothing with the CFP.

Another example of the statistical or qualitative reproducibility is the famous d - d fusion reactions at low energy as discussed by Huizenga in his book (equations are numbered as in the book);

"D + D→ [⁴ He]* → ³ He (0.82 MeV) + n (2.45 MeV),	(3.8.2a)
$\rightarrow t (1.01 \text{ MeV}) + p (3.02 \text{ MeV}),$	(3.8.2b)
\rightarrow ⁴ He (0.08 MeV) + γ (23.77 MeV).	(3.8.2c)

The reactions (3.8.2a) and (3.8.2b) have been studied over a range of deuteron kinetic energies down to a few kilo-electron volts (keV) and the cross sections (production rates) for these two reactions have been found experimentally to be nearly equal (to within ten percent). Hence, the fusion of deuterium produces approximately equal yields of 2.45 million-electron-volts (MeV) neutrons (with an accompanying ³He atom) and 3.02–MeV protons (with an accompanying tritium atom). This near-equality of the neutron and proton branches (production rates) is expected also on the basis of theoretical arguments. The cross section (production rate) for reaction (3.8.2c) is several orders of magnitude lower than reactions (3.8.2a) and (3.8.2b)." [Huizenga 1992 (pp. 6–7)]. (Numbers of the equations are renumbered at citation.)

The fusion reaction of two deuterons with energies down to a few keV occurs with probabilities for three channels given in Eqs. (3.8.2) as explained in the above sentence by Huizenga [Huizenga 1992]. If the results are averaged over many events, then we will obtain the products according to the probabilities determined by the branching ratios. The individual product, however, shows an unexpected value not described by the probability in a short term measurement where we observe only few reactions. This is another example of the qualitative (or probabilistic) reproducibility in nuclear physics where it is usual laws in microscopic processes.

It should be noticed another phase of truth in the sentence by Huizenga cited above. We know a doubt expressed by M. Fleischmann in his first paper on the mechanism of the CFP;

"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] (The reactions (v) and (vi) in this sentence correspond to reactions (3.8.2a) and (3.8.2b) written above in this paper.)

The point we want to notice is the different reactions of Huizenga and Fleischmann to the experimental fact: Huizenga pointed out only the contradiction between the fact and the scheme of existing science while Fleischmann noticed something new in the same contradiction as Huizenga noticed.

Here, we remember a parable about our recognition told by ancient Chinese saint: "When you are angry, you cannot be correct. When you are frightened, you cannot be correct; when there is something you desire, you cannot be correct; when there is something you are anxious about, you cannot be correct. When the mind is not present, we look, but do not see. We listen, but do not hear; we eat, but don't taste our food. This is the meaning of "the cultivation of the person lies in the correction of the mind." [Great Learning (9. The cultivation of the person lies in the correction of the mind.)]

We see that the cause of the difference in the responses of two scientists to the same fact is based on the desire they had in their mined; "*when there is something you desire, you cannot be correct;*" From my point of view, the desire in the mind of Huizenga disturbed his sight into the truth through the experimental facts.

Evidence of stochastic occurrence of events (at least the emission of neutrons) in the CFP is clearly shown in Fig. 3.8.1 by an excellent experiment by Gozzi et al. [Gozzi 1991]. As is well known, the Poisson distribution is a discrete probability distribution that expresses the probability of a given number of events occurring in a fixed interval of time and/or space if these events occur with a known average rate and independently of the time since the last event. One of examples in physics that may follow a Poisson is the number of decay events per second from a radioactive source, as cited above the alpha-decay of ${}^{226}_{88}$ Ra.



Fig. 3.8.1 Frequency count of neutrons as observed in 5421 intervals of ten minutes acquisitions and as expected in a Poisson distribution. The variability of the expected values obtained allowing the measured mean value to vary between $\mu - \sigma = 0.32$ and $\mu + \sigma = 0.37$ counts/10 min. is also reported [Gozzi 1991 (Fig. 13)].

3.8.3.4 Self-organization and Chaotic Behavior of a Microscopic State in Non-equilibrium Condition beyond Control by Macroscopic Conditions

As we have discussed already in a paper published in 2013 [Kozima 2013], there is a possibility that the optimum microscopic state in a CF material composed of a host element and a hydrogen isotope, e.g. the superlattice made of a sublattice of the host element and another sublattice of the hydrogen isotope, is generated by self-organization in the non-equilibrium CF materials. It is, of course, the process governed by nonlinear dynamics and is not controllable macroscopically. This characteristic is discussed by many researchers in terms of the complexity as cited below:

"The constructive role of irreversibility is even more striking in far-from-equilibrium situations where non-equilibrium leads to new forms of coherence." [Prigogine 1996 (p. 26)]

"Nonequilibrium leads to concepts such as – self-organization and dissipative structures, - - - ." [Prigogine 1996 (p.27)]

"Could unpredictability itself be measured? The answer to this question lay in a Russian conception, the Lyapunov exponent. This number provided a measure of just the topological qualities that corresponded to such concepts as unpredictability. The Lyapunov exponents in a system provided a way of measuring the conflicting effects of stretching, contracting, and folding in the phase space of an attractor. They gave a picture of all the properties of a system that lead to stability or instability. An exponent greater than zero meant stretching—nearby points would separate. An exponent smaller than zero meant contraction (stability). For a fixed-point attractor, all the Lyapunov exponents were negative, since the direction of pull was inward toward the final steady state. An attractor in the form of a periodic orbit had one exponent of exactly zero and other exponents that were negative. A strange attractor (chaos), it turned out, had to have at least one positive Lyapunov exponent." [Gleick 1987 (p. 253)]

The stability of a system is determined by the sign of the Lyapunov exponent of the system described by a difference equation (cf. Section 3.8.3.2). As explained in our paper [Kozima 2013], the Feigenbaum's theorem tells us that various kinds of systems obeying a single hump distribution of the recursion function show the chaotic behavior, and therefore unpredictability or irreproducibility.

3.9 Neutrons in Transition-Metal Hydrides and Deuterides

As have been demonstrated above by reviewing the typical experimental data on the CFP, we have to accept the fact that there are nuclear reactions in CF materials at near room temperature. This fact is out of common sense in nuclear physics developed mainly in 20th century. Then, what is the hidden parameter for the occurrence of nuclear reactions in CF materials not recognized in nuclear physics where the fields of research have been confined for nuclei in free space except a short time when they interact each other. One of remained fields not considered in ordinary nuclear physics is the quasi-stable state of neutrons in solids. Our model was based on this point [Kozima 2015 (10), 2016a].

We have developed a quantum mechanical explanation of the premises assumed in the TNCF model, especially the existence of the trapped neutrons in such CF materials as NiH_x , PdD_x , carbon graphite, cross-linked polyethylene (XLPE). In the investigation, we noticed new knowledge of nuclear and solid state physics in their frontiers developing rapidly.

It is interesting to recollect the frontiers of nuclear physics at 1960s when the author was a graduate student. The late Professor Toshinosuke Muto (1904 – 1973), Tokyo University, my instructor at the graduate course, used to say at seminars on nuclear physics that there remain many interesting research themes in the low energy nuclear physics (energy region below a few MeV) while the high energy region (more than 100 MeV) was attracting many researchers at that time. His comment on the importance of low and medium energy nuclear physics at that time has shown its correctness when we notice the recent vivid researches in ultra-low energy neutrons and exotic nuclei in terms of the shell model [Kozima 2014c, 2016a]. A new trend of recent researches in nuclear physics is expressed in the following sentence by Sahin et al.: "The interplay among central, spin-orbit, and tensor components of the effective nucleon-nucleon interaction can shift effective single-particle energies relative to each other as protons and neutrons fill certain orbitals near the Fermi surface in nuclei with large neutron excess." [Sahin 2015].

Therefore, we have to say that the physics of CF materials (the system composed of lattice nuclei and hydrogen isotopes, such as NiHx, PdDx, CHx, etc.) where the lattice nuclei and interstitial hydrogen isotopes are interacting through the nuclear force and the lattice nuclei each other are interacting with the super-nuclear interaction mediated by hydrogen isotopes is in its infantile stage [Kozima 2006, 2014c]. Experimentally, the CFP and the diffusion characteristics of hydrogen in transition metals might be subtle signals of the new states of neutrons in CF materials. Theoretically, the new knowledge of the exotic nuclei on one hand and the characteristics of transition-metal hydrates on

the other are the hint to cultivate the physics of CF materials [Kozima 2016a].

The exotic nuclei with an excess of neutron numbers such as ${}^{11}_{3}$ Li and ${}^{12}_{4}$ Be are the examples of them suggesting the possible exotic nuclei participating in the CFP such as ${}^{A}_{6}$ C, ${}^{A}_{28}$ Ni, and ${}^{A}_{46}$ Pd as discussed before in terms of the TNCF model [Kozima 2014b, 2014c]. The recent works on the exotic nuclei have confirmed existence of ${}^{32}_{12}$ Mg [Utsuno 2014], ${}^{42}_{14}$ Si [Stroberg 2014], ${}^{69}_{29}$ Cu [Morfouace 2014], ${}^{73}_{29}$ Cu [Sahin 2015], ${}^{92}_{42}$ Mo [Sharp 2013] and they have been investigated in relation to the bases of the shell model of nucleus as discussed in another paper [Kozima 2014c].

Our first attempt to approach the quasi-stable neutrons in solids is an attempt to formulate neutron energy bands formed by the neutron-lattice interaction similar to the electron energy bands well known in solid state physics [Kozima 1998a (Sec. 12.4), 1998c, 2006 (Sec. 3.7.2.2)]. The idea of the neutron energy bands has recently been developed as presented at JCF16 [Kozima 2016a].

Even if the investigation of neutron physics participating in the CFP developed in the papers cited above is a tentative one, the treatment may be a first step to the neutron physics in solids, which we hope to play a role corresponding to the historical liquid-drop model of nuclei proposed by N. Bohr for nuclear reactions in the early stage of nuclear physics to describe a global feature of the CFP and the physics of neutrons in CF materials.

4. Conclusion – The Cold Fusion Phenomenon as a Science of Neutrons in Solids

Consistent understanding of the whole events in the CFP needs the Science of the CF materials composed of host elements and hydrogens (protium and/or deuterium) such as NiH_x, PdD_x, CH_x, and so forth.

The typical papers on the CFP introduced in this paper show clearly that this phenomenon includes events with wide variety occurring in various CF materials composed of various host elements and hydrogen isotopes (both protium and deuterium).

As we have shown in our books and papers including this paper [Kozima 1998a, 2006, 2014a, 2014b, 2016b, 2016c], it is possible to give a unified explanation of the whole experimental data on the CFP using our TNCF model based on the phenomenological approach where we assumed existence of trapped neutrons in the superlattice composed of a sublattice of the host element and another sublattice of the hydrogen isotope formed in CF materials. There appears, then, a possibility to have the super-nuclear interaction between lattice nuclei mediated by interstitial hydrogen

isotopes and the formation of neutron bands where are "the trapped neutrons" assumed in the TNCF model. The trapped neutrons form the CF matter where are formed neutron drops ${}^{A}_{Z}\Delta$ similar to the neutron-proton clusters figured out in the neutron star matter by Negele et al. [Negele 1973]. The CF matter, then, participates the nuclear reactions in the CF materials observed as excess heat and nuclear transmutations in the CFP.

Thus, we can conclude according to the success of the phenomenological approach that the science of the CFP is the science of neutrons in solids which have never noticed its existence until a part of its finger tips was revealed by the work published in 1989 by Fleischmann et al. [Fleischmann 1989].

Appendix: Episode of Excess Heat Measurement in Protium System

Pioneers in this unknown field of the CFP have experienced unexpected facts and they had to work by a trial-and-error approach. In doing so, they made many mistakes caused by mismatching between their presumptions based on the common sense they had and the facts they observed in their experiments. This is a normal process in pioneering works in science even if the discrepancy in the CFP was rather enormous.

The two books by G. Taubes [Taubes 1992] and J.R. Huizenga [Huizenga 1992] had cited many examples of these mistakes performed in the early days of the CF research and gave negative influence against the CFP to people who did not investigate concrete facts.

An episode about the control experiment by F. Pons is taken up by both opponents in their books as if the heat measurement by Fleischmann et al. is incredible. We can read their paragraphs telling the episode below. Paragraphs from Huizenga [Huizenga 1992] and from Taubes [Taubes 1992] are given with underlines in corresponding parts at citation.

"Furth, in an excellent review paper on nuclear fusion, discussed progress toward achievement of practical fusion power. He was the token nuclear physicist speaking at the Dallas ACS session. In his talk, Furth discussed also the extremely small probabilities of fusing hydrogen isotopes at room temperature and the large effective electron mass that would be required to account for the University of Utah claims. Furth concluded that many additional experiments needed to be performed before nuclear physicists would believe the University of Utah's reported data. One of the crucial experiments he suggested was to compare light water (H₂O) and heavy water (D₂O) water under the same electrolytic conditions. Pons replied that he was preparing to do this. On the other hand, based on the discussion following Pons' lecture at Dallas it appeared that <u>Pons and Fleischmann had already performed this control experiment.</u> When Pons was asked why he had not reported results of control experiments with light water substituted for heavy water, he replied "A baseline reaction run with light water is not necessarily a good baseline reaction." When asked to elaborate, Pons intimated he had performed the experiment with light water and had seen fusion, saying "We do not get the expected baseline experiment. . . We do not get the total blank experiment we expected" (Science 244, p. 285)." [Huizenga 1992, pp. 31 – 32] (The underlines are added at citation.)

"The collaboration, however, was undone by Pons's feelings of persecution and then by the local lawyers. First, Linford had a run-in with Pons, sparked by a slight Pons felt he had suffered at the congressional hearings. Harold Furth of Princeton had called Linford before the hearings to learn exactly what Pons had said about his light water controls in his Los Alamos seminar. Linford, who had a videotape of the seminar, found the point at which Pons answered the question about light water – that he had seen heat and then discontinued the experiment – and played it for Furth over the telephone. In Washington, Furth had apparently confronted Pons with what he had said in Los Alamos, suggesting it was proof cold fusion did not exist. Pons had not taken it well. Now Linford stopped by the Utah lab as Pons and Fleischmann were showing the Texas A&M people around; then he slipped away for a few minutes with Pons." [Taubes 1992, pp. 260 – 261] (The underlines are added at citation.)

The citation of this episode by Huizenga and Taubes given above shows their clear intention to show how incredible is the excess heat measurements by CF researchers and to denunciate the discovery of the CFP. However, regrettably to them, the history of the CF research in these more than 25 years have shown reality of the CFP and occurrence of unexpected events in hydrated and deuterated solids revealing a realm of new physics unknown before. Really, great discoveries are often found by such unexpected observations as many examples show in the history of science as X-ray by W.C. Roentgen (1895) and radioactivity by A.H. Becquerel (1896). [Kozima 1998a (Appendix D)]. It is said that "*Becquerel's discovery of spontaneous radioactivity is a famous example of serendipity of how chance favors the prepared mind*." (Wikipedia [Henri Becquerel]).

It is possible to imagine according to the episode cited above that the first observation of the CFP in the protium systems, which is now recognized by almost all CF researchers, was performed by Pons and his collaborators in the University of Utah before or in the year of 1989.

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