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Cold Fusion Phenomenon in the Composite CF Materials – Mixed Hydrogen Isotopes, Alloys, Ceramics, and Polymers –⁺

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Abstract

Using the TNCF (trapped neutron catalyzed fusion) and ND (neutron drop) models, which were successful to give a unified explanation of various kinds of experimental data sets obtained in a great variety of CF materials hitherto, we explained various features of the cold fusion phenomenon (CFP) observed in the composite (multi-component) CF materials, CF materials composed of host elements (alloys, ceramics, and polymers) and hydrogen isotopes (H or/and D). We take up in this paper following CF materials: (1) Alloys, (2) Ceramics, and (3) Polymers including XLPE and biological systems in addition to (4) the case where used a mixture of hydrogen isotopes, H and D.

Despite of the rather complex host materials of various compositions and structures in the composite CF materials, we could give a consistent explanation of the specific experimental data obtained in them. We have proposed a tentative criterion for a minor element added to a major element (Pd or Ni) to make the alloy to be a composite CF material. Because of the importance of the composite CF materials used frequently in recent works with many interesting results especially on the improved qualitative reproducibility and the elevated amount of excess heat generation, it is useful to understand the physics of the nuclear reactions occurring there. The fundamental problems related to the premises of our models in relation to the composite CF materials will be discussed in another paper presented in this Conference.

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References

1. Introduction

In the papers presented until JCF19 (December 2018), we took up mainly the “simple” cold fusion materials (CF materials) composed of a simple host element and a hydrogen isotope and analyzed experimental data sets including various events in the cold fusion phenomenon (CFP) with the inductive logic using phenomenological approach. The reason of such an approach with the inductive logic is justified as experimental results obtained in various CF materials are effects of the same cause satisfying the common necessary conditions: One of the necessary conditions should be the formation of the superlattice of host elements and hydrogen isotopes. This is the reason of applicability of the meta-analysis as discussed in a recent paper [Kozima 2019a].

We investigate the experimental data obtained in the composite and the compound CF materials using the phenomenological approach used for the CF materials with simple host elements and show our approach is effective also to the rather complex systems.

1.1 Composite and Compound CF materials

The two kinds of CF materials listed below had been left out of our detailed investigation because their complex structure perplexed us. They are (1) the composite materials including alloys, ceramics and polymers and (2) the compound materials including various kinds of interfaces between two CF materials, between a CF material and its substrate, or between a CF material and its environment (cf. [Kozima 2021a (Introduction)]).

It should be noticed that the interfaces have been inevitable parts of any CF material and they are classified into four types: the solid-solid, solid-liquid, solid-gas and solid-plasma interfaces. The second type has been investigated most extensively in the electrochemistry [Bockris 1970a, 1970b, 2000, Horiuti 1951, 1970, Kita 1971, 1973] but other types are also important in the CFP. It is, however, probable that the characteristics of the solid-liquid interface investigated thoroughly in the electrochemistry are common, at least partially, to other interfaces and are very important in our research on the CFP in the composite and compound CF materials. This problem will be investigated specifically in the forthcoming paper [Kozima 2021c].

In the investigation of the CFP in the group (2) listed above (the compound CF materials), all interfaces belonging to the four types may influence the CFP in these CF materials. The first group (the composite CF materials) is taken up in this paper and the second group (the compound CF materials) will be investigated in another paper presented in this Conference [Kozima 2021a].

It is possible to say that these materials (1) and (2) pointed out above have been investigated by many researchers from rather the application point of view to find out effective candidates for application than the scientific one to clarify the mechanism realizing the CFP.

The CFP is, in our opinion, realized in materials (CF materials) where are formed a superlattice of host elements and hydrogen isotopes (H or D) [Kozima 2006, 2021b]. The hydrogen isotopes for the formation of the superlattice are supplied from outside through the interfaces around the host material to feed protons/deuterons by chemical reactions at around solid electrodes in the electrolytic system (cf. e.g. [Bockris 1970a, 1970b, 2000, Horiuti 1970]) including the so-called “*hydrogen electrode reaction* (HER)” [Horiuti 1951, Kita 1971, 1973] where the catalytic action plays an essential role, which will be discussed in detail in another paper [Kozima 2021c].

In the case of the CFP in the composite CF materials, the supply of the protons/deuterons to the host CF material noticed in the above paragraph seems to have close relation with the “*active sites*” of catalysis and the “*supported catalysis*” defined as follows:

“A heterogeneous catalyst has **active sites**, which are the atoms or crystal faces where the reaction actually occurs. Depending on the mechanism, the active site may be either a planar exposed metal surface, a crystal edge with imperfect metal valence or a complicated combination of the two.” [Wikipedia Catalysis]

“Heterogeneous catalysts are typically **supported**,” which means that the catalyst is dispersed on a second material that enhances the effectiveness or minimizes their cost.” [Wikipedia Catalysis]

In this paper, we will introduce many examples of the CFP in the composite CF materials from experimental data sets in the CFP and suggest the role of the *active sites* and the *supported catalysis*.

Some features of the CFP related to the HER at the interfaces will be discussed in another paper on the CFP in the compound CF materials presented at this Conference [Kozima 2021a].

1.2 Necessary Conditions for the Cold Fusion Phenomenon (CFP)

The necessary conditions for the CFP are expressed as follows: Formation of CF materials = Realization of the superlattice of host elements and hydrogen isotopes (PdD, NiH, etc.) \leftarrow Occlusion of H/D in the host materials \leftarrow Hydrogen electrode reaction ($2\text{H}^+ + 2\text{e}^- \rightleftharpoons \text{H}_2$) at the interface of the host material = A kind of catalysis.

Main features of these flow are taken up in several papers presented by us:

- (1) Explanation of essential features of the CFP by a phenomenological approach i.e. the TNCF model and the Neutron Drop model [Kozima 1998, 2006, 2021b (Introduction)].
- (2) Explanation of various phases of the CFP by the TNCF and the TN models in the phenomenological approach [Kozima 1998, 2006, 2019b].
- (3) General investigation of the effects of the catalytic action on the CFP will be given elsewhere [Kozima 2021c]

There are several kinds of CF materials which we did not take up specifically until now. They include as pointed out above two kinds of materials; (1) the *composite materials* including alloys, ceramics, and polymers and (2) the *compound materials* with interfaces between several parts (materials supported by substrates, multi-layered materials, etc.) composing the system . The materials in the second group are (a) CF materials surrounded by liquids and gases, (b) those supported by substrates, (c) and those have multilayered structure.

In this paper, we discuss the CFP in the CF materials in the group (1) and the CFP in the group (2) will be discussed in another paper presented in this Conference [Kozima 2021a].

1.3 Short History of Our Research on the CFP

In the analyses presented until JCF19 (December 2018), we took up whole experimental data sets as tabulated in Table 1.1 below. We have employed in the inductive logic using the phenomenological approach using a model named the TNCF (trapped neutron catalyzed fusion) model to investigate the various events in the CFP observed in a variety of materials as results of the same cause satisfying the necessary conditions such as the formation of the superlattice of host elements and hydrogen isotopes (protons or deuterons) on the basic assumption of applicability of the meta-analysis [Kozima 2019a].

It should be emphasized that the partner of host elements for the superlattice is either protons or deuterons. The necessary and sufficient condition for the CFP is the formation of the neutron bands containing the trapped neutrons (neutron Bloch waves) in them. The meta-analysis has shown that the trapped neutron induces the CFP independent of the species of the hydrogen isotope in the CF material if the superlattice is formed.

Table 1.1. Systems and Obtained Evidence of the CFP: Host solids, agents, experimental methods, direct and indirect evidence, cumulative and dissipative observables are tabulated. Q and NT express excess energy and the nuclear transmutation, respectively. The direct evidence of nuclear reactions in the CFP is the dependences of reaction products on their energy (ϵ) and position (r), the decrease of decay constants of radioactive nuclides, the decrease of fission threshold energy of compound nuclei.

Host solids	C, Pd, Ti, Ni, Au, Pt, KCl + LiCl, ReBa ₂ Cu ₃ O ₇ , Na _x WO ₃ , KD ₂ PO ₄ , TGS (triglycine sulfate), SrCe _a Y _b Nb _c O _d , XLPE (cross linked polyethylene), Biological Systems (microbial cultures) Pd _x Ni _{0.35-_x} Zr _{0.65} ,
Agents	$n, d, p, {}^6_3\text{Li}, {}^{10}_3\text{B}, {}^{12}_6\text{C}, {}^{39}_{19}\text{K}, {}^{85}_{37}\text{Rb}, {}^{87}_{37}\text{Rb}$
Experiments	Electrolysis, Liquid contact, Gas discharge, Gas contact
Direct evidence of nuclear reaction	Gamma ray spectrum $\gamma(\epsilon)$, Neutron energy spectrum $n(\epsilon)$, Space distribution of NT products NT(r), Stabilization of unstable nuclei (Decrease of decay constants), Lowering of fission threshold energy
Indirect evidence of	Excess energy Q , Number of neutrons N_n , Amounts of tritium atom N_t , Helium-4 atom* N_{He4} , NT products (NT _D , NT _F , NT _A), X-ray

nuclear reaction	spectrum $X(\varepsilon)$
Cumulative observables	$NT(\mathbf{r})$, Amount of tritium atom N_t , Amount of helium 4* N_{He4} ,
Dissipative observables	Excess energy Q , Neutron energy spectrum $n(\varepsilon)$, Number of neutrons N_n , Gamma ray spectrum $\gamma(\varepsilon)$, X-ray spectrum $X(\varepsilon)$,

(In the rows of “Direct evidence” and “Indirect evidence,” observables relevant to discussions given in this paper are printed in red.)

On the other hand, the investigation of the events taken up in this and the next papers [Kozima 2021a] is rather deductive than inductive used in the research in the former works (cf. [Kozima 2019a]). The case studies, so to speak, of experimental data here depend on individual conditions in the experiments asking appropriate explanation for their relationship with the necessary conditions developed in the inductive research based on the meta-analysis of a lot of positive experimental data. Especially important in this paper is the chemical characteristics of such host transition elements as Ti, Ni, Pd, Pt, etc. showing subtle but magical catalytic action at interfaces with their environment. The “supports” noticed in the heterogeneous catalysis [Wikipedia Catalysis] may have a close relation with the CFP in the composite CF materials taken up in this paper. An extensive investigation of this problem will be given in another paper [Kozima 2021c] but a brief glimpse of possible influence of the supports will be discussed in this paper.

The physical applicability of our models, the original TNCF and the Neutron Drop models [Kozima 2006], is explained briefly in another paper presented at this Conference [Kozima 2021a]. The subtle problem of the catalysis in relation to the generation of the necessary conditions for the CFP will be investigated extensively elsewhere.

1.4 Nuclear Reactions in Free Space and in the CF Materials

It should be noticed the fundamental difference of nuclear reactions generating nuclear transmutations in the CF materials from that in free space, first.

1.4.1 Nuclear reactions in free space

Reactions in free space supposed by Fleischmann et al. to be responsible to their experimental results are written down as follows [Fleischmann 1989, Kozima 2019c]:

$$d + d \rightarrow {}^4_2\text{He}^* \rightarrow t (1.01 \text{ MeV}) + p (3.12 \text{ MeV}), \quad (1.1)$$

$$\rightarrow {}^3_2\text{He} (0.82 \text{ MeV}) + n (2.45 \text{ MeV}), \quad (1.2)$$

$$\rightarrow {}^4_2\text{He} (0.08 \text{ MeV}) + \gamma (23.8 \text{ MeV}). \quad (1.3)$$

The branching ratios of these reactions have been determined in the nuclear physics as $1 : 1 : 10^{-7}$ down to low energies of a few keV.

For the convenience of our discussion in this paper, we cite nuclear reactions between light nuclei. Reactions in free space related to the TNCF model. [Kozima 1997]

$$n + p = d (1.3 \text{ keV}) + \gamma (2.2 \text{ MeV}). \quad (1.4)$$

$$n + d = t (6.98 \text{ keV}) + \gamma (6.25 \text{ MeV}). \quad (1.5)$$

$$n + {}^6_3\text{Li} = t (2.7 \text{ MeV}) + {}^4_2\text{He} (2.1 \text{ MeV}), \quad (1.6)$$

$$t + d = {}^4_2\text{He} (3.5 \text{ MeV}) + n (14.1 \text{ MeV}), \quad (1.7)$$

$$n + d = n + p + n, \quad (1.8)$$

$$\gamma + d = p + n. \quad (1.9)$$

Unified Explanation of the CFP by the Phenomenological Theory using models – the TNCF model and the Neutron Drop Model – is given by us in which the gamma in Eqs. (1.4) and (1.5) in free space should be read as φ 's (phonons excited by the reaction in contact with trapped neutrons) as explained below. [Kozima 1994, 1997, 1998, 2000, 2006, 2019a, 2019b]

1.4.2 Nuclear reactions in CF materials

The phenomenological theory using the trapped neutron catalyzed fusion model (TNCF model) [Kozima 1994] has shown its effectiveness to give a unified consistent explanation of the experimental data sets with full of variety in the kinds of the CF materials and in the observed physical quantities [Kozima 1998, 2006]. The trapped neutrons with a density n_n , one of the key premises of the model [Kozima 1994, 1998] has been extended to the neutrons in the neutron energy bands formed by the super-nuclear interaction between neutrons in lattice nuclei in the superlattice made of host elements and hydrogen isotopes [Kozima 2006]. The name “the trapped neutron” will be used throughout this paper to call the neutrons in the original meaning in the early papers [Kozima 1994, 1998] and the neutrons in the neutron energy bands in the recent papers [Kozima 2006, 2019b]. The trapped neutrons in the neutron bands and the itinerant protons/deuterons form the *cf-matter* containing the trapped neutrons and the neutron drops ${}^A_Z\Delta$, composed of Z protons and $(A - Z)$ neutrons, in the CF material where the superlattice is formed [Kozima 2005, 2006 (Sec. 2.4.2)].

The brief explanation of the TNCF model is given in the books [Kozima 1998, 2006] and papers [Kozima 2005, 2021b (Appendix A2)].

According to the discussions for the phenomenological approach given in these books

and papers, we have assumed that the gammas in the reactions (1.4) and (1.5) in free space should be read as φ 's (phonons excited in the CF materials by the fusion reactions in contact with trapped neutrons) in the CF material:

$$n + p = d + \varphi's + Q, \quad (1.4')$$

$$n + d = t + \varphi's + Q', \quad (1.5')$$

where $Q = 2.2$ MeV and $Q' = 6.25$ MeV.

It is interesting to notice the liberated energies Q_{n-p} and Q_{n-d} by and the fusion cross sections σ_{n-p} and σ_{n-d} for the reactions (1.4) and (1.5). The liberated energies are $Q_{n-p} = 2.2$ MeV and $Q_{n-d} = 6.25$ MeV, respectively. And the fusion cross sections for the reactions σ_{n-p} and σ_{n-d} in free space are given as 3.22×10^{-1} and 5.5×10^{-4} barns, respectively.

$$\sigma_{n-p} = 3.22 \times 10^{-1} \text{ barns}, \quad (1.10)$$

$$\sigma_{n-d} = 5.5 \times 10^{-4} \text{ barns}. \quad (1.11)$$

These values will be used in the discussion of the experiments by Fleischmann et al. [Fleischmann 1989] given in Sec. 2.4.1.

1.5 Fundamental Problems related to the Formation of the CF Materials

In this investigation, we must take care of the subtle effect of the catalytic nature of the key host elements such as Pd, Ni and Ti on the formation of the CF materials especially at its first stage of adsorption of protons/deuterons on the interface. This factor of the key elements has been investigated in the electrochemistry for long but not carefully in the cold fusion experiments yet and its detailed consideration will be given in near future [Kozima 2021c].

1.6 Characteristics of the Nuclear Reactions in the CFP

“A remarkable characteristic of the nuclear reactions in the CFP is the fact that there are observed no gamma radiations accompanied to the nuclear reactions producing transmuted nuclei in contrast to the case in free space where are observed always any radiation such as gamma, beta and alphas with energies liberated by the nuclear reaction [Kozima 1998, 2006, 2017]. Furthermore, there is another remarkable characteristic of the nuclear transmutations in the CFP that the generation of new elements A_ZX with large shifts of the mass number A and the proton number Z from those of elements pre-existed in the CF materials. It is also noticed that the observed distribution of A_ZX over the proton number Z is in accordance with the abundances of elements in the universe [Kozima 2006, 2012b, Suess 1956, Trimble 1975].” [Kozima 2019d (Table 2.1)]

(1) Relations between Observed Values of Physical Quantities. [Kozima 1998 (p. 150),

2006 (p. 76)]

$$N_Q \approx N_t \approx 10^7 N_n. \quad (1.13)$$

- (2) Surface nature of nuclear reactions in the CFP [Kozima 1998 (Sec. 12.16), 2006 (Sec. 2.2.1.), 2011a]

Summary of the analyses of the experimental data by the TNCF model is given in Appendix A1. Recognition of the value of the phenomenological approach using the TNCF model is traced historically in Appendix A2. It is interesting to notice that very few of CF researchers expressed their favor to the phenomenological approach as seen in Appendix A2.

1.7 Outline of This Paper

In this paper, we explain various features of the cold fusion phenomenon (CFP) observed in the composite (multi-component) CF materials, i.e. CF materials composed of host elements (alloys and ceramics, etc.) and hydrogen isotopes (H or/and D). In our phenomenological approach, we used the TNCF (trapped neutron catalyzed fusion) and ND (neutron drop) models, which have been successful to give a unified explanation of various kinds of experimental data sets obtained in a great variety of CF materials hitherto, are applied to deduce explanation of the specific experimental data obtained in these composite CF materials with various compositions and structures. We take up in this paper following CF materials: (1) Alloys, (2) ceramics, and (3) polymers including XLPE and biological systems. The characteristics of the CFP observed in these composite CF materials have been explained successfully by our model as we have done for the various experimental data obtained in rather simple host materials as published hitherto.

Several examples of the composite CF materials taken up in this paper are (a) Pd cathodes with an electrolytic solution 0.1 M LiOD in 99.5% D₂O + 0.5% H₂O solutions [Fleischmann 1989], (b) Stainless steel cathode Fe_{1-x-y}Cr_xNi_y [Dufour 1993], (c) Pd-Rh alloys, PdRh_xCo_yB (x=5%, y=3%), PdRh_xCr_y (x=5%, y=5%) [Claytor 1998], (d) Ceramic cathode TiC, VC, ZrB, ZrC, ZrN, LaB in glow discharges [Romodanov 1998b], (e) Stainless steel cathode Fe₇₁Cr₁₈Ni₁₀Ti₁ in glow discharges [Romodanov 1998c], (f) Nickel alloy Ni_{7.6}Cr_{20.6}Fe_{70.4}Mn_{1.4} in gas contact experiments [Campari 2004b], (g) Constantan Cu₅₅Ni₄₄Mn₁ [Celani 2012]. (h) Pd-Ni alloys Pd_xNi_{0.35-x}Zr_{0.65} [Kitamura 2018].

Various features of the experimental results obtained in these composite CF materials have been explained from our point of view briefly summarized in the beginning of this abstract.

2. Brief Review of the Fundamental Works in Relation to the Composite CF Materials

2.1 Superlattice of the Host Element and the Hydrogen Isotope

The formation of the superlattice XH/XD of a host element X and a hydrogen isotope H/D had been investigated as a process in complexity and out of our control resulting in the qualitative reproducibility, a characteristic of the CFP [Kozima 2013]. The specific attention about the effect of the minor element A_ZX added to a host element Ni or Pd on the CFP will be discussed in another paper presented in this Conference [Kozima 2021b].

2.2 TNCF and Neutron Drop Models for the Cold Fusion Phenomenon

To give a unified explanation for the very many experimental data sets obtained in various systems with variety of components of host elements and either protium and/or deuterium, we proposed a phenomenological approach based on a model (TNCF model) [Kozima 1994, 1998, 2006, 2021a]. Fortunately, we could give a unified qualitative, even sometimes semi-quantitative, explanation for the whole data obtained in this field. Details have been presented in the papers and books cited above and references given there.

2.3 Effects of Background Neutrons

It should be emphasized that the experimental data on the effects of thermal neutrons on the realization of the CFP, even if we described them in our papers and books already.

We can have no CFP without background neutrons [Jones 1994, Forsly 1998, Kozima 1998]. We can cite a convincing sentence by S. Jones describing this fact as follows: “We invite those with evidence for neutron production to accept our invitation to test their system **in the deep-underground neutron detection facility in Provo Canyon to confirm results**. Gamma and x-ray spectrometers are also available on request.” ([Jones 1994 (p. 26-13)] [Kozima 1998 (Sec. 8.1)])

On the contrary, there are many evidence showing a positive effect of thermal neutrons on the CFP [Shani 1989, Yuhimchuk 1992, Celani 1992, Stella 1993, Oya 1996, Lipson 1996] [Kozima 1998 (Sec. 8.2)].

2.4 The CFP in the CF Materials with Composite Host Elements

The analyses of many experimental results using the TNCF model had been published in many papers and the results were compiled in our two books published in 1998 [Kozima 1998] and 2006 [Kozima 2006].

Several splendid ones of very many experimental results would be referred as follows [Fleischmann 1989, Jones 1989, Packham 1989, Morrey 1990, Srinivasan 1990, McKubre 1993, Ohmori 2016] which had been analyzed and presented in our books and papers. Detailed discussion on the peculiarity of the composite CF materials will be given in Section 4 from our present point of view.

2.4.1. On the Mixed Hydrogen Isotopes in Electrolytic Solution

There are several experiments where used solvent including both light and heavy waters for the electrolyte. We introduce here only two experiments by Fleischmann et al. [Fleischmann 1989] and Swartz et al. [Swartz 2003, 2019] which are interesting from our point of view.

2.4.1.1 Experiments by Fleischmann et al. [Fleischmann 1989]

We want to add a word on the experiment by Fleischmann et al. [Fleischmann 1989] where they used an electrolytic solution, 0.1 M LiOD in a solvent of mixed light and heavy water (99.5% D₂O + 0.5% H₂O), as explained in their sentence cited below:

“In the work reported here D⁺ was compressed galvanostatically into sheet, rod and cube samples of Pd from 0.1 M LiOD in 99.5% D₂O + 0.5% H₂O solutions. Electrode potentials were measured with respect to a Pd-D reference electrode charged to the α-β-phase equilibrium. We report here experiments of several kinds:” [Fleischmann 1989 (p. 302)]

Why did they add “0.5% H₂O” to the electrolytic solution? This fact is not considered seriously hitherto but the addition of H₂O may be closely related to the large difference about three orders of magnitudes of the fusion cross sections of the above reactions (1.4) and (1.5), $\sigma_{n-p} = 3.22 \times 10^{-1}$ and $\sigma_{n-d} = 5.5 \times 10^{-4}$ barns, respectively. The reaction (1.4) will heat the CF material in this case PdD_x/PdH₂ to suffice necessary conditions for the CFP related to the temperature effect [Kozima 2020].

From our point of view, the PdD_x system forms an alloy stable for large ranges of deuteron contents x and the CFP in this system is explained by the self-organization of the superlattice PdD in the open, non-equilibrium condition [Kozima 2013].

If this speculation is correct, we may be able to depict a following explanation: It is probable that the process of occlusion of hydrogen isotopes through the solid-liquid

interface depends on the species of the isotopes as we know from knowledge of electrochemistry. However, we may assume that the amounts of deuterons and protons included in the CF material are around $n_d : n_p = 99.5 : 0.5$ in ratio, close to the ratio of $D_2O : H_2O$ in the solution.

Then, we can anticipate that the addition of 0.5% H_2O to the electrolytic solution do not influence much on the formation of the neutron energy bands (and therefore the generation of the cf-matter) but influence strongly on the reaction between the trapped neutron (neutrons in the neutron band) and hydrogen isotopes *at disordered situation* of the protons by the following speculation.

In the formation of the super-nuclear interaction between two neutrons in different lattice nuclei mediated by interstitial protons/deuterons, the effects of the different species of the interstitial hydrogen isotopes are averaged out. While the interaction between a neutron Bloch wave and a proton/deuteron as a two-body collision reflects directly the disordered situation of the latter; the position and the difference of species. A neutron Bloch wave encounters with protons more often than with deuterons in this case.

Neglecting this factor, we can estimate the large effect of the addition of 0.5 % H_2O in the solvent on the system as follows.

Let us estimate the numbers of the two-body $n-p$ and $n-d$ collisions simply as two body collisions between free particles (neglecting the enhancement of the $n-p$ interaction due to the disordered situation of the protons) generating excess energies $Q_{n-p} = 2.2$ MeV and $Q_{n-d} = 6.25$ MeV, respectively. Using the absorption cross-section determined in the Nuclear Physics, we obtain the ratio of excess heat generations by $n-d$ and $n-p$ reactions,

$$\begin{aligned} \sigma_{n-d} n_d Q_{n-d} : \sigma_{n-p} n_p Q_{n-p} &\approx 5.5 \times 10^{-4} \times 99.5 \times 6.25 : 3.22 \times 10^{-1} \times 0.5 \times 2.2 \\ &= 5.47 \times 6.25 : 37.2 \times 2.2 \\ &= 34.2 : 81.8. \end{aligned}$$

This ratio shows that the contribution of the excess heat generation by the $n-p$ reaction is almost three times larger than that by the $n-d$ reaction for the elevation of the sample to induce the temperature effect on the nuclear reactions between the trapped neutrons and the deuterons resulting in the excess heat generation in the system [Kozima 2013 (Sec. 3.1), 2020].

2.4.1.2 Experiments by Swartz et al. [Swartz 2003, 2019]

The solvent used by Swartz et al. are opposite to that used by Fleischmann et al. They used solvents composed of light water containing small amount of heavy water up to 3.7 % with Ni cathode and Pt anode.

The characteristic of their experiments is the effect of the heavy water added to the light

water solvent on the excess energy evolution. Their results are qualitatively explained as follows:

“Heavy water (D₂O) yields significant increases in the excess heat observed for nickel light water systems for all input electrical power levels examined (250 to 1500 milliwatts).”

[Swartz 2003 (p. 335, Abstract)]

“These results indicate a loss of deuterium (more precisely, “deuterons”) from ordinary water when excess heat is observed in an aqueous Ni CF/LANR system (Ni/ordinaryH₂O/Pt) using a very large cathodic area.” [Swartz 2019 (p. 169, Abstract)]

From our point of view, the NiH_x system forms an alloy stable for large ranges of hydrogen contents x and the CFP in this system is explained by the self-organization of the superlattice NiH in the open, non-equilibrium condition [Kozima 2013]. From this point of view, the experimental results obtained by Swartz et al. are explained as follows.

Fundamentally, the NiH superstructure is generated by self-organization when there are formed NiH_{1-x}D_x ($x < 0.04$) alloys by the diffusion of H and D from the solid-liquid interface on the Ni surface. The super-nuclear interaction between neutrons in lattice Ni nuclei results in the neutron bands and the neutrons in the bands (the trapped neutrons) make nuclear interactions with disordered nuclei including protons and deuterons (cf. Sec. 1.4). There should be other nuclear interactions of the trapped neutron with disordered nuclei but we write down only those with a proton (at disordered position) and a deuteron:

$$n + p = d + \varphi's + Q, \quad (1.4')$$

$$n + d = t + \varphi's + Q', \quad (1.5')$$

where $Q = 2.2$ MeV and $Q' = 6.25$ MeV.

The deuterons are fundamentally exotic nuclei in the superlattice NiH and the interaction (1.5') occurs more frequently than (1.4'). This is the reason Swartz et al. observed much excess heat with more deuterons [Swartz 2003] and loss of deuterium when excess heat is observed [Swartz 2019].

2.4.2 Cold Fusion Phenomenon in the Composite CF Materials

The experimental data sets performed with the composite CF materials, alloys, ceramics and polymers, have been piled up in these almost 30 years and show various characteristics difficult to give quantitative explanation due mainly to the complicated structure of their samples. We introduce them qualitatively in Section 4 after summarization of fundamental natures of the nuclear reactions in the CF materials.

3. Chemical and Physical Conditions on the CF Materials

It is our regret that we did not consider the chemical conditions in the CFP even if we touched slightly on them in relation to the Pd-D-Li and Ni-H-K combinations as favorable ones for the realization of the CFP [Kozima 2004].

In the reality of the CFP, the chemical conditions are very important for the realization of the necessary conditions for the nuclear reactions especially in the earlier stages of the superlattice formation as the absorption and occlusion of hydrogen isotopes depend strongly on the catalytic action at interfaces [Kita 1971, 1973, Wikipedia Catalysis].

We give a brief investigation of this problem in Section 3.1 leaving detailed treatments on this problem to the following paper [Kozima 2021c].

3.1 Effects of Electrolyte in the Electrolytic Experiments

In the early days of the research on the CFP, the effects of electrolytes and impurities in electrolytic experiments had been investigated [McBreen 1990].

“The absorption of hydrogen and deuterium in Pd, at potentials positive to hydrogen evolution, was studied by cyclic voltammetry. Results in 0.1 M NaOH, LiOH and LiOD were compared. In LiOH hydrogen ingress into Pd is faster than in NaOH. However, hydrogen egress is inhibited. In LiOD all processes are slower than those in LiOH.” [McBreen 1990 (p. 279, Abstract)]

It was shown that addition of a very small quantity of cyanide into the electrolytic solution had resulted in several major effects on the behavior of the electrolysis.

“Addition of 10^{-3} M CN^- to these electrolytes resulted in several major effects. The formation of the oxide and the ingress of hydrogen and deuterium were inhibited. Egress of hydrogen and deuterium from Pd was essentially completely inhibited until the adsorbed cyanide was displaced by oxide at positive potentials. Hydrogen permeation measurements through Pd membranes were made in 0.1 M NaOH and in 0.1 M NaOH + 10^{-3} M NaCN. Cyanide adsorption decreased the permeation rate and had large effects on the permeation decay transients.” [McBreen 1990 (p. 279, Abstract)]

Ulmann et al. had shown the large effects of contamination of a Pd cathode on the electrolysis.

“One of the typical characteristics of the hydrogen evolution reaction is its extreme sensitivity to various impurities present in the solution. - - - We showed recently [3] that the contamination of a palladium cathode, polarized in LiOD + D_2O solution, with lead and more so with zinc leads to the buildup of very large over voltages for D_2 evolution. In the present communication we describe the results of surface analyses for a series of Pd electrodes used as cathodes in the D_2O electrolysis cells run for up to 34 days.”

[Ulmann 1990 (p. 257, Abstract)]

[3] J. Augustynski, M. Ulmann and J. Liu, *Chimia*, **43**, p. 355 (1989). [Augustynski 1989]

“We then ask how hydrogen reacts at the interface with the metal, whether and how hydrogen diffuses into the metal and forms a solid solution, a metal hydride or other types of ‘hydrogen-metal-alloys’.” [Schlapbach 1991 (p. 409, Introduction)]

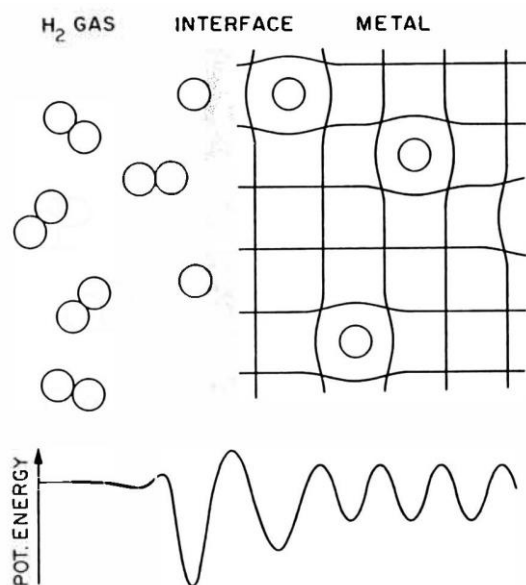


Fig. 3.1. Simplified model of the dissociation of molecular hydrogen at an interface and of the solution of hydrogen atoms in the bulk, on interstitial sites.” [Schlapbach 1991 (p. 410, Fig. 1)]

*“There is experimental and theoretical evidence that chemisorbed H does not necessarily occupy sites on top of the first metal atom layer, but also **sites between and underneath top surface metal atoms**. Subsurface H was observed together with a strong surface reconstruction (surface hydride formation).”* [Schlapbach 1991 (p. 415)]

3.2 Temperature Effects (Appendix A2)

It should be remembered that the temperature of the sample has decisive influence on the occurrence of the CFP as several experimental data had shown it clearly.

We determined a quantitative measure of the temperature effect on the CFP in the paper published in 2008 [Kozima 2008].

“1) If the temperature of the experimental system is lower than a critical temperature T_c (from the experiment, we may take it higher than 60 °C; $T_c \geq 60$ °C), there are no cold

fusion reactions.

2) Increasing the temperature higher than T_c ($T_c < 90\text{ }^\circ\text{C}$ as confirmed by experiments), the cold fusion reactions start to occur and produce excess energy inducing more reactions as we see in the occurrence of many pulses.

This means that the critical temperature T_c in this case is in between 60 and $90\text{ }^\circ\text{C}$ ($60\text{ }^\circ\text{C} \leq T_c \leq 90\text{ }^\circ\text{C}$).

3) We may assume that higher temperature is more favorable for the CFP if other conditions are the same. Then, when the temperature of the sample increases due to CF reactions generating more excess energy than dissipating energy from the system and the shape of the tubular cathode is favorable of the positive feedback of the CFP, there may occur a positive feedback which increases the temperature of the system, thus inducing more CF reactions.” [Kozima 2008]

The larger increase of the diffusivity in NiH than in PdH(D) may be responsible to the more often occurrence of nuclear transmutations in NiH than in PdD tabulated in figures in our books ([Kozima 1998 (Figs. 11.2 and 11.3) and 2006 (Figs. 2.22 and 2.3)])

It is also discussed in relation to the diffusivity of H/D as follows:

“It is well known that the diffusivity of hydrogen isotopes in transition metal hydrides/deuterides is very large and depends exponentially on the temperature [15, 16]. As shown in Figs. 1 and 2, the diffusion coefficient of H in Pd increases almost two orders of magnitude in the temperature range 0 and 200 °C and that of H in Ni almost three orders of magnitude in the temperature range 25 and 300°C [15].” ([Kozima 2020])

([15] [McKubre 1993] and [16] [Yamaguchi 1993])

Heating CF material by chemical reactions (crystallization, chemical reaction, etc.) results in the elevation of the temperature of the sample and accordingly in acceleration of nuclear reactions in the CFP. This effect seems to be observed in the recent experiments by Kitamura et al. [Kitamura 2018] as pointed in our recent paper [Kozima 2020].

[Reifenschweiler 1995]

“A sharp decrease of the radioactivity of tritium was observed when the hydrogen isotope is sorbed by small monocrystalline particles of titanium and the preparation is heated to several hundred degrees centigrade.” [Reifenschweiler 1995 (p. 163)]

[McKubre 1993]

“Using a different calorimetric system, a higher than first order dependence has been

reported previously⁶ for the variation of excess power with electrochemical current. An important distinction between **this previous study** and **the results reported here** is that, in the former case, the calorimetric system was not operated, as **here, at constant total input power with an efficient heat removal mechanism**, thereby ensuring an approximately constant mean cell temperature. Instead, the system was operated at constant electrochemical current **in a heat-retaining calorimeter design**, thereby forcing the mean cell temperature to increase with increasing current. Clearly, further work is required to elucidate the nature of this apparent temperature dependence of the excess power.” [McKubre 1993 (p. 17)]

[Claytor 1991]

“In typical operation, a unipolar, square pulse with a width of at least 150 μ s at a repetition rate of 100 pulses per second was used at voltages as high as possible before breakdown occurred, typically 1200 to 2500 V. Currents of up to 5 A were used in some experiments.” [Claytor 1991 (p. 400)]

Without loss of energy fed to the 8-layer Pd powder, the temperature of the sample increases up to 4.35 to 54.4 K in a second.

3.3 Effects of Chemistry at Interfaces and the Qualitative Reproducibility

It is inevitable to have some kinds of interfaces surrounding the CF material (necessarily solid) where we observe the CFP. The interfaces are divided into four; (1) solid-solid, (2) solid-liquid, (3) solid-gas, and (4) solid-plasma interfaces. The chemistry of atomic processes at the solid-liquid interface have been extensively investigated for more than 70 years in the electrochemistry and many interesting characteristics of chemical reactions, especially the HER (hydrogen electrode reaction) [Bockris 1970a, 1970b, 2000, Horiuti 1951, 1970].

Especially interesting is the dependence of catalytic reactions on the lattice plane facing to the interface [Kita 1971, 1973]. It had been well known fact that the catalytic reaction is subtle about the existence of minute additives for long and the data given by Kita et al. added a concrete evidence to it.

This knowledge obtained in the electrochemistry on the solid-liquid interface may have common characteristics for the chemistry of atomic reactions at other interfaces; solid-solid, solid-gas and solid-plasma interfaces. Then, the subtleness of the catalytic action depending on the polycrystalline structure of the solid at the interfaces, which is

not controllable macroscopically, gives rise to the qualitative reproducibility of the CFP depending strongly on the inclusion of hydrogen isotopes through the interfaces surrounding the CF material.

We have now two causes of the qualitative reproducibility, one pointed out before the *self-organization of the superlattice* in the CF material by a process of complexity, and another pointed out above the catalytic action depending on *the polycrystalline structure of the CF material*.

Now, it is possible to say that the nature of the CFP explained by our phenomenological approach shows the qualitative reproducibility is a nature of this research field and should not be rejected by the lack of the quantitative reproducibility existing only in physics where works only linear interactions in it. The problem of the reproducibility of the events in the CFP had been one of tumbling blocks for acceptance of this field by the established science community for more than 30 years without correct knowledge of the science of the CFP.

4. Experimental Data in the Composite CF Materials – Alloys, Ceramics and Polymers (XLPE and Biological Systems)

There are many experimental data obtained in the CF materials with the composite structure, that is alloys, ceramics and polymers composed of several kinds of host elements.

[McKubre 1993]

*“As a possible solution to this problem, we have observed that **the addition of small amounts (typically 200 ppm) of non-classical additives, such as aluminium or silicon (in metallic and oxide form, respectively), to the electrolyte, results in the ability to maintain high loadings for longer periods, without impeding the initial attainment of high loadings. In this context, it should be mentioned that, in cells which utilize glass components, silicon-containing species will accumulate in basic electrolytes over extended time periods.**”* [McKubre 1993 (p. 6)]

4.1 Alloys.

In the early days of the research on the CFP, many researchers have taken up alloys as CF materials guided perhaps by their properties of large occlusion and/or of large diffusivity of hydrogen isotopes in them. In the period when there have been no guiding principles to choose host materials other than their properties in relation to the hydrogen

isotopes noticed above, the above mentioned standard of the choice is a reasonable one and revealed several characteristics for the realization of the nuclear reactions in the CF materials at around room temperature. Some features of these characteristics have been introduced in our papers and books [Kozima 1998 (Sec. 6.1e,), 2006 (Section 2.2.2)].

In these more than 30 years after the discovery of the CFP in 1989, there have accumulated experimental data performed in composite CF materials. We would like to classify tentatively these CF materials into two groups; (1) Alloys and ceramics containing at least one element which is known as a host element of a CF material showing the CFP, and (2) Alloys and ceramics composed of several elements which have not been known as a host element of a CF material by now.

The first group, let us name them as **Type I alloys and ceramics**, includes Stainless Steel ($\text{Fe}_{71}\text{Cr}_{18}\text{Ni}_{10}\text{Ti}_1$), Pd alloys used by Claytor et al. (such as PdRh_x and PdCu_x), Constantan ($\text{Cu}_{55}\text{Ni}_{44}\text{Mn}_1$) used by Celani et al., Ni alloys used by Campari et al. ($\text{Ni}_{7.6}\text{Cr}_{20.6}\text{Fe}_{70.4}\text{Mn}_{1.4}$), Ni alloys used by Iwamura et al. ($\text{Ni}_x\text{Cu}_y\text{Ca}_z$) and Pd/Ni alloys used by Kitamura et al. ($\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$). In this type I CF materials, we may be able to include the original CF materials composed of single elements Ti, Ni, Pd, Pt, Au and others.

The second group, let us name them as **Type II alloys and ceramics**, includes ceramics used by Romodanov et al. (such as VC, ZrB, ZrC, ZrN, LaB) and those used by Mizuno et al. (such as SrCeO_3 and $\text{Sr}_{1.0}\text{Ce}_{0.9}\text{Y}_{0.08}\text{Nb}_{0.02}\text{O}_{2.97}$). In this type II CF materials, we may be able to include the graphite which had shown a specific CFP of various nuclear transmutations [Kozima 2012a, 2017 (Sec. 2.1.1)].

The classification of the CF materials into two types, Type I and Type II, will be investigated in our forthcoming paper in relation to the characteristics of the hydrogen diffusivity in them [Kozima 2021d].

Alloys and ceramics cultivated for the CF materials are heat-resisting in general and the CFP in them have been observed in the higher temperature region up to 1000 °C (up to 3000 °C in the case of graphite) utilizing the temperature effect for the super-nuclear interaction in the CF material [Kozima 2008, 2013 (Sec. 3.1)].

Experiments on alloys and ceramics performed at elevated temperatures are discussed in Appendix A2 in relation to the temperature dependence of diffusivity of H/D in these CF materials.

4.1.1 Stainless Steel by Sparking in Hydrogen Isotopes [Dufour 1993]

Dufour et al. have measured excess heat generation in stainless steel occluding hydrogen isotopes with the discharge experiment [Dufour 1993]. It is well known that SS

is composed of Cr-Fe-Ni with following compositions:

$^{A}_{24}\text{Cr}$ ($A = 50, 52, 53, 54$)

$^{A}_{26}\text{Fe}$ ($A = 54, 56, 57, 58$)

$^{A}_{28}\text{Ni}$ ($A = 58, 60, 61, 62, 64$)

We cite their reports as follows:

“Sparking in hydrogen isotopes (H_2 and D_2) between electrodes of various metals (palladium, stainless steel) yields, in the setup we have used, an excess energy production that is fully reproducible and stable over very long periods. No systematic errors have been found that could explain the statistically significant excess energy production we have measured in comparison with reference experiments”. [Dufour 1993 (p. 227)]

“Chromium Cr exhibits extremely low solubility for hydrogen even though a hexagonal monohydride CrH and a cubic dihydride CrH₂ are reported [Shavely 1949]. There are no CFP data in chromium hydrides and deuterides. However, $^{53}_{24}\text{Cr}$ (9.55 %) and $^{57}_{26}\text{Fe}$ (2.19 %), and $^{54}_{24}\text{Cr}$ (2.38 %) and $^{58}_{26}\text{Fe}$ (0.33 %) can absorb a deuteron ($\Delta A = 2$), and a proton ($\Delta A = 1$), respectively to produce stable Mn, and Co isotopes. Therefore, the stainless steels $\text{Fe}_{1-x-y}\text{Cr}_x\text{Ni}_y$, widely used in technology, may be responsible for CFP at high temperatures where hydrogen isotopes can permeate through them. Experimental data of CFP in the stainless steel with D_2 and H_2 in discharge experiments are reported by Dufour [Dufour 1993].” [Kozima 2006 (p. 99)]

4.1.2 Ceramics and Stainless Steel in Discharge Experiments [Romodanov 1998]

Romodanov et al. had performed elaborate experiments on the CFP with the solid-plasma interfaces in the discharge with various cathodes including several metals, ceramics, and alloys [Romodanov 1998a, 1998b, 1998c]. A part of their data sets had been introduced in our books [Kozima 1998 (Sec. 6.4e), 2006 (p. 99)]. Experiments with ceramics will be treated in Sec. 4.2.

Ceramics and Alloys

Ceramics; TiC, VC, ZrB, ZrC, ZrN, LaB [Romodanov 1998b].

Metals and Alloys; Fe, Alloy SS, Ti, Nb [Romodanov 1998c] (SS; Fe-Cr-Ni-Ti (71-18-10-1)).

In the glow discharge experiments, they observed tritium generation in several ceramics:

“4.1 The significant tritium generation rate, is fixed at radiation by ions deuterium of

samples from borides, carbides and nitrides of titanium, vanadium, zirconium and lanthanum from plasma powerful of glow discharge. For samples of ZrB₂, the tritium generation rate reached $(3.6 - 3.9) \times 10^7$ atom/s.

4.3 *The most proof of materials, at radiation in plasma of glow discharge, were ZrB₂ and ZrN, and the least proof samples of LaB₆.*” [Romodanov 1998b (p. 594, Conclusion)]

They had resistive heater to heat up the samples up to $T = 670\text{K} - 1200\text{K}$ (900 and 1100 – 1200 K for SS) and made them contact with H₂, D₂, H₂ + D₂ at $P = (5 - 60) \cdot 10^3$ Pa (for 0.1 – 50 h). [Romodanov 1998c].

“The research is conducted on samples of iron and its alloys as well as on samples of titanium and niobium at interaction, basically, with ordinary hydrogen. It is established that the short-term bursts of tritium generation arising at interaction of hydrogen isotopes in which is immersed, previously heated up metal sample, relate to output tritium, early accumulated, owing of effects isotopes.” [Romodanov 1998c (p. 319, Abstract)]

It should be added a data by Romodanov et al. [Romodanov 1998c] that tritium had been generated in stainless steel heated by electric coil. [Kozima 2006 (p. 99)]

4.1.3 Pd Alloys [Claytor 1998]

Claytor et al. in the Los Alamos National Laboratory had made experiments on the various types of Pd alloys. We summarize their experiments on the following Pd alloys in this paper and will give more extensive review of their papers on various types of CF materials in another paper to be presented at this Conference [Kozima 2021a (Appendix A3)].

The experimental data we take up in this section is on the following CF materials: PdRh_xCo_yB ($x=5\%$, $y=3\%$), PdRh_x ($x=10, 5, 0.5, 0.1\%$), PdCu_x ($x=1, 10\%$), PdFe_x ($x=10\%$), PdRh_xCr_y ($x=5\%$, $y=5\%$), PdCr_x (1.1%), PdNi_x (1.1%)” [Claytor 1998 (p. 90)]

They observed production of tritium in several alloys and investigated effects of minor elements in the CF materials on the production rate of tritium.

“Most of the alloys described in this paper were made by a melt spinning (produces a tape) process. A boron nitride crucible was used to contain the molten palladium alloys and it cannot be excluded that nearly all the melt spun alloys contain some boron as an unintentional impurity. Several of the alloys were made in a batch process by arc melting powder on a cooled copper block then homogenizing the alloys and subsequently rolling them into thin (220 microns) foils without intermediate annealing. Alloy concentrations

are all given in weight percent.” [Claytor 1998 (p. 89)]

“Most of the alloys did not produce tritium, only the PdRhCoB, PdCu, PdRh (0.1%) and PdB gave significant upward slopes. Surprisingly, several of the alloys, PdLi, PdBe, and PdHf produced distinctly negative slopes. It is unknown, at present, if this is due to actual tritium removal or a suppression of the background ion current.” [Claytor 1998 (pp. 90 – 91)]

Table 4-1. Palladium alloys and highest tritium rates observed during the plasma experiments (a part of Table 1 in [Claytor 1998]).

alloy composition	alloy type	Number of runs	highest tritium rate, nCi/h	significance
PdRh (10%)	melt spun	1	-0.00073	1
PdRh(5%)	melt spun	1	-0.0024	1
PdRh (0.5%)	rolled alloy	1	-0.00017	0
PdRh (0.1%)	homogenized, rolled alloy	1	+0.0048	2

“Note that the Rhodium series (PdRh_x ; $x=10, 5, 0.5, 0.1\%$) of alloys gives a negative slope at high concentrations and a positive slope at lower concentrations. This same effect was found before in technical grade palladium, in that increasing the amount of impurities in the palladium is likely to suppress the tritium output. However, it appears that the copper (PdCu_x ; $x=1, 10\%$) and boron $\text{PdRh}_x\text{Co}_y\text{B}$; $x=5\%, y=3\%$) alloys at higher concentrations produced higher tritium output rates. More data should be collected in these two alloy systems to confirm these interesting results.” [Claytor 1998 (pp. 90 – 91)]

“The critical factor in the tritium yield is the purity and metallurgical condition of palladium. Alloys have been discovered that gave much enhanced tritium output over randomly selected materials. Unfortunately, not all the conditions necessary for a completely reproducible experiment are known at this time. The results presented here are encouraging in that it appears that designed materials may be fabricated that will reproducibly generate tritium and perhaps show other anomalous effects at measurable levels.” [Claytor 1998 (p. 92, Conclusion)]

This experimental results on the impurity dependence of the tritium production are very interesting in relation to the analysis of the data by Fleischmann et al. [Fleischmann 1989] given in Sec. 2.4.1. As we have given justification of our model to the composite

CF materials [Kozima 2021b], the addition of some minor elements (*CF active elements*) to the host material (Pd in this case) may not give fatal influence for the generation of the cf-matter including neutrons and deuterons. (This situation has similarity to the case of Fleischmann et al. where a small amount of H₂O added to D₂O will not influence much for the generation of the cf-matter).

The reasoning of this speculation may be described as follows in the case of the host alloy Pd_{1-x}X_x when the element X is a CF active one:

(1) The first effect appears in the formation of the cf-matter: In the super-nuclear interaction in an alloy Pd_{1-x}X_xD ($x < 10\%$), the nuclear interactions Pd-*d* and X-*d* between an interstitial deuteron and the surrounding lattice nuclei Pd or X are summed up and finally the resulting super-nuclear interaction between neutrons in different lattice nuclei does not suffer much from the addition of foreign element X when its concentration is small. (cf. the similar reasoning in the case of Pd + (99.5 % D₂O and 0.5 % H₂O) in Sec. 2.4.1).

(2) The second effect appears in the interaction between the trapped neutron and other nuclei: The nuclear reactions between a trapped neutron (a neutron in a neutron energy band) and nucleus A_ZX in disordered sites such as written down below suffer directly by the addition of foreign nuclei which the trapped neutron sees as disordered nuclei:

$$n + {}^A_ZX = {}^{A+1}_ZX^* = {}^{A+1}_ZX + \phi's, \quad (4.1)$$

$$= {}^{A+1-a}_{Z-b}X' + {}^a_bX'' + \phi's, \quad (4.2)$$

including the tritium generating reaction

$$n + d = t + \phi's. \quad (1.5')$$

On the other hand, the addition of a CF nonadditive

General Tendency: When the concentration of the foreign element X is small, the effect (1) does not influence the cf-matter formation and the effect (2) is observed which increases with the concentration of X.

Dependence on the Species of the Minor Element.

When the concentration of X (Rh) increases, the effects (1) gives the deterioration of the cf-matter and the nuclear reactions between the trapped neutron and other nuclei (including deuterons) decrease in the case of PdRh_x ($x=10, 5, 0.5, 0.1\%$). This story gives explanation of the data given in Table 1.

In the case of PdCu_x ($x=1, 10\%$) and PdRh_xCo_yB ($x=5\%, y=3\%$) alloys, on the other hand, the effect (1) by X (Cu, Co) is small and X intensifies the effect (2) and also the reaction (1.5'). This is an explanation of the fact observed in these alloys [Claytor 1998 (pp. 90 – 91)].

This may be the cause of the events observed by Claytor et al. [Claytor 1998] described in the paragraph cited above (with emphases marked by red).

It should be necessary to add a word on the detection of tritium in the CFP. As we see in our papers presented at this Conference and others presented former Conferences, the detection of the tritium has been a most fundamental and reliable evidence of the nuclear reactions from 1989 and up to now. One of the first experimental results confirmed the generation of the tritium was that by Packham et al. reported in 1989 [Packham 1989]. The reliability of the experimental data in such a science as the CFP should be judged from the meta-analytic point of view as we had shown in the paper presented at JCF19 [Kozima 2019a]. This viewpoint is confirmed also in this paper where cited several experimental results showing qualitative coincidence of tritium and neutron observation.

4.1.4 CF Active and CF Inactive Elements [Claytor 1998]

From the experimental data obtained by Claytor et al. [Claytor 1998], we must care about the species of the minor elements in the composite CF materials to extend the phenomenological approach to apply them due to the following reason.

- (1) It is revealed by Claytor et al. [Claytor 1998] that addition of minor elements to a host element (Pd, for instance) causes the increase of tritium generation by some element (B and Cu, for instance) and its decrease by some others (Li, Be, and Hf, for instance).
- (2) On the other hand, there are several evidence that iron-group elements are active in the HER (hydrogen electrode reaction) [Kita 1973] and UPD (underpotential deposition) [Akiyama 1986, Fukushima 1993, Nakano 1998, Bockris 2000 (Sec. 7.12.11)].

Considering the experimental data given above (1) and (2), we may be able to define tentatively (a) CF active (or constructive) elements and (b) CF inactive (or destructive) elements as follows according to the inductive logic [Kozima 2019a]:

- (a) *CF active elements*; elements in 3d and 4d transition metals.
- (b) *CF inactive elements*; other elements than those in the group (a).

Then, we may be able to use the analogy of the electron bands in the (In, Ga)N alloy to the neutron bands in the CF materials composed of Pd (or Ni) and *CF active elements* as discussed in our paper presented at this Conference [Kozima 2021b (Appendix A6-2)].

Fortunately, according to the above criterion, the TNCF model is applicable to these CF materials including those used in recent works by Celani et al. (Constantan $\text{Cu}_{55}\text{Ni}_{44}\text{Mn}_1$) and Kitamura et al. ($\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$).

4.1.5 Ni Alloys $\text{Ni}_x\text{Cu}_y\text{Ca}_z$ and $\text{Ni}_x\text{Cu}_y\text{Y}_z$ [Iwamura 2020a, 2020b]

Iwamura et al. had performed extensive experiments with a specific CF materials composed of the so-called the Pd complexes (Pd/CaO/Pd) and (Pd/MgO/Pd) through which permeated the D_2 gas (cf. [Kozima 2021a (Sec. 4.4.5)]). Recently, they made an interesting experiments of excess heat and elemental distributions on the “nano-sized multilayer metal composite” samples composed of Ni, Cu, CaO or Y_2O_3 shown in Fig. 4.1 (In the figure caption, elements of the layers are colored corresponding to that in the figure).

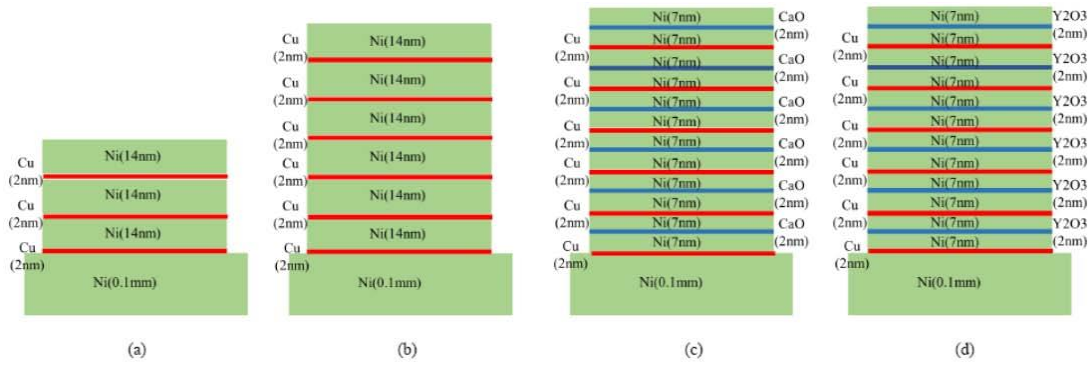


Fig. 4.1. Structures of nano-sized multilayer metal composite samples; (a) example #1: 3 layers of **Cu** (2 nm) and **Ni** (14 nm), (b) Example #2: 6 layers of **Cu** (2 nm) and **Ni** (14 nm), (c) Example #3: 6 layers of **CaO** (2 nm), **Cu** (2 nm) and **Ni** (14 nm), (d) Example #4: 6 layers of **Y₂O₃** (2 nm), **Cu** (2 nm) and **Ni** (14 nm). [Iwamura 2020b (Fig. 4)]

The magnitudes and temporal variations of the excess heat generation in the composite sample with different compositions reveal interesting characteristics depending on the composition and the temperature of the samples as shown in Fig. 4.2. (In the figure caption, materials of the layers are colored corresponding to the figure.)

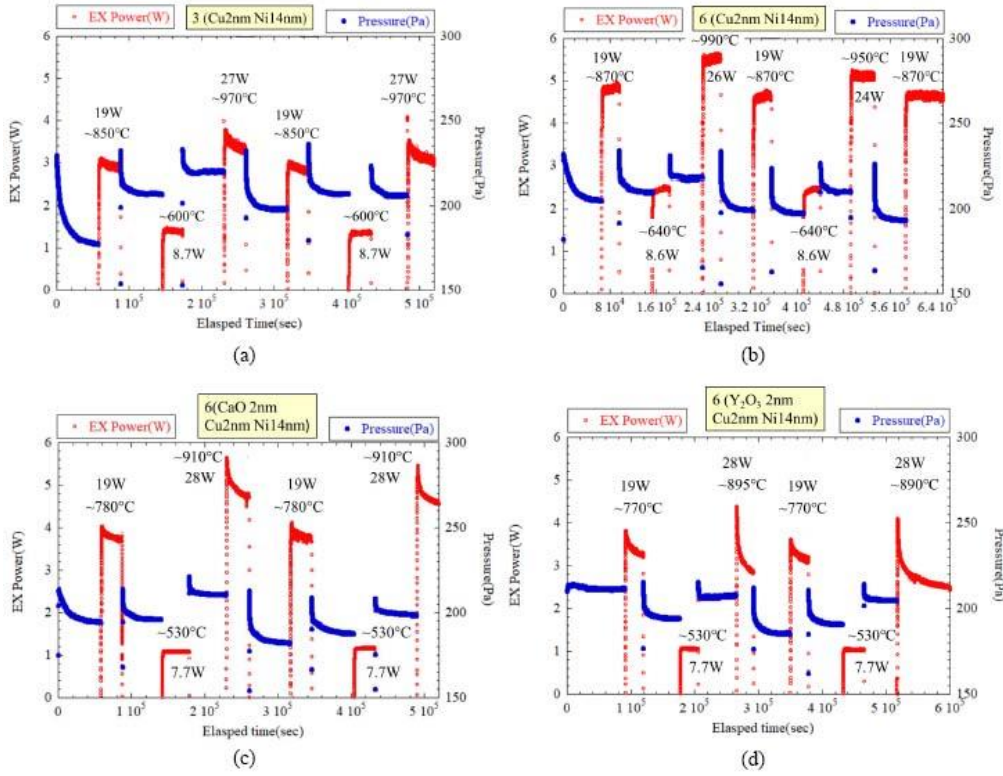


Fig. 4.2. Examples on excess heat generation; (a) example #1: 3 layers of Cu (2 nm) and Ni (14 nm), (b) example #2: 6 layers of Cu (2 nm) and Ni (14 nm), (c) example #3: 6 layers of CaO (2 nm), Cu (2 nm) and Ni (14 nm), (d) example #4: 6 layers of Y₂O₃ (2 nm), Cu (2 nm) and Ni (14 nm). (The powers and temperatures written in the figure (19 W, ~850°C in (a), for example, show the input power to and the temperature of the sample at the beginning of the excess heat generation.) [Iwamura 2020b (Fig. 5)]

It is possible to give our interpretation for the amounts and the temporal dependences of the excess heat given in Fig. 4.2 as follows.

Firstly, we notice a characteristic of alloys for the CFP, existence of active and inactive minor elements as explained in Sec. 4.1.4 above and more extensively in another paper presented at this Conference [Kozima 2021b (Appendix A6-2)]. According to this classification, Cu is an active element which assists or intensifies the CFP when added to the main element (Ni in this case) in the CF material. According to the experimental data by Iwamura et al. [Iwamura 2020b] we can classify Ca in active and Y in inactive elements (which work in opposite direction for the CFP) as discussed below in the item (7). The activity of Cu for the CFP is qualitatively understood looking into the values of diffusivity of hydrogen isotopes in Pd, Ni and Cu shown in Appendix A3 in this paper.

Secondly, the experimental data on the depth profiles of elements are shown in Fig. 3 for a sample (six layers of Cu (2 nm) and Ni (14 nm) with Y₂O₃ corresponding to the data (d) in Fig. 4.2) after excess heat generation. The data in Fig. 4.3 (b) show that the layer structure of the sample almost disappeared finally after the series of the excess heat generation in 6×10^5 sec. This means that the sample transformed into the state of alloys composed of elements in the original layers after the nuclear reactions generating the excess heat in 167 h.

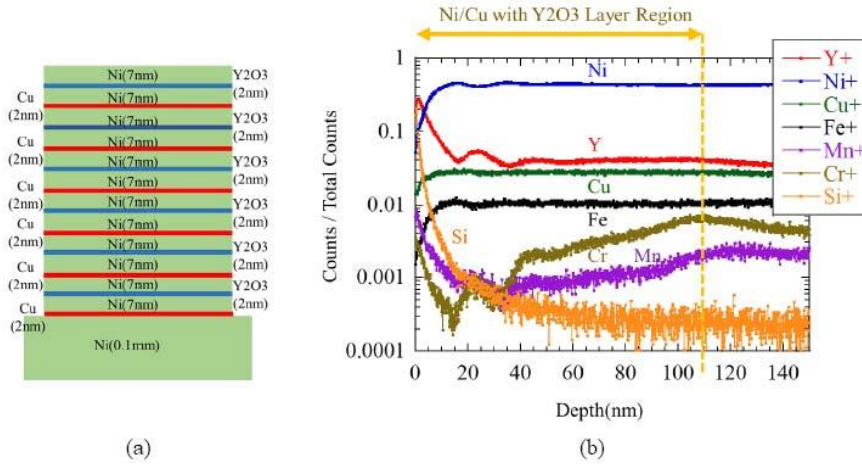


Fig. 4.3. An example of TOF-SIMS analysis; (a) structure of the sample before experiment, (b) depth profiles of the after the series of the excess heat generations. [Iwamura 2020b (Fig. 11)]

Then, we can give a story telling us the nuclear reactions and the structure changes occurring in the samples depicted in the Figs. 4.2 and 4.3 as follows.

- (1) The NiH superlattice is formed near the solid-gas interface of the sample.
- (2) The neutron energy bands and therefore the cf-matter including the trapped neutrons are generated where the superlattice is formed.
- (3) The trapped neutrons induce nuclear reactions between protons ($n - p$ reactions) and other nuclei A_ZX 's ($n - {}^A_ZX$ reactions) at disordered sites; main sites may be near the solid-solid interfaces of the layered structure. The difference of the excess heat generation in Figs. 4.2 (a) and (b) may be explained by the number of solid-solid interfaces 3 in (a) and 6 in (b); the excess heat in (a) is almost 60 % of that in (b) while the ratio of the layers is 0.5.
- (4) The main reaction participating in the excess heat generation in the experiments may be the following n - p fusion reaction:

$$n + p = d + \varphi's + Q, \quad (4.3)$$

where $Q = 2.2$ MeV and the fusion cross-section is $\sigma_{n-p} = 3.22 \times 10^{-1}$ barns.

- (5) The liberated excess energy by the reaction (4.3) (and if any other) heats up the sample and destroys finally the layer structure of the sample (Fig. 4.3 (a)) into alloys as shown in Fig. 4.3 (b).
- (6) In the case of alloys composed of Ni and Cu (active element), the cf-matter formed by the trapped neutrons and itinerant protons remains without much damage and the reaction (4.3) last long as appeared in Fig. 4. 2 (a) and partially in (b). The increase of the excess power seen in Fig. 4.2 (b) might be the enhancing effect of the active impurity observed by Claytor et al. [Claytor 1998] introduced in Section 4.1.3.
- (7) In the case of other alloys including Ca or Y as minor elements, the cf-matter may be influence by the characteristics of the minor elements. Looking into the data in Fig. 4.2 (c) and (d), we see that Ca is an active element for the CFP in Ni and Y is inactive if we can ignore other factors influencing to the nuclear reactions in the experiments.
- (8) The temperature effect on the CFP [Kozima 2008, 2013 (Sec. 3.1), 2020 (Sec. 2-(3))], the higher the sample temperature the more the nuclear reactions, is clearly shown in the excess heat vs. heater temperature relation as seen in Fig. 4.2 and as shown more clearly in Fig. 4.4 [Iwamura 2020b (Fig. 6)].

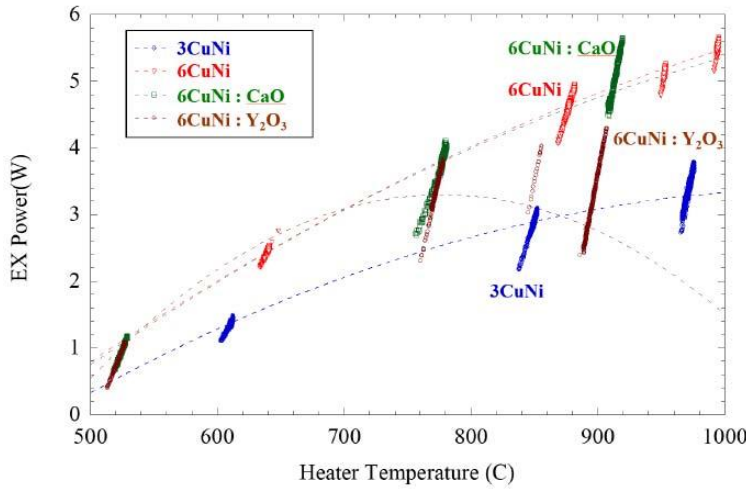


Fig. 4.4. Temperature dependence on excess power for four examples. [Iwamura 2020b (Fig. 6)]

It is interesting to notice that the temporal decreases of the excess heat generation do not appear in Fig. 4.2 (b) at all and in Figs. 4.2 (a), (b) and (c) when the input powers to the sample are less than 9 W in (a) and 8 W in (c) and (d).

4.1.6 Ni Alloys $\text{Ni}_x\text{Cr}_y\text{Fe}_z\text{Mn}_{1.4}$ ($x = 7.6$, $y = 20.6$, $z = 70.4$) [Campari 2004a, 2004b]

Campari et al. [Campari 2004a, 2004b] investigated the CFP in the CF materials of the Ni-H system where observed emissions of photons and neutrons along the heat production at high temperature in the range of 400 to 700 K. They performed investigation of surface transformation of the samples where observed the above CF effects. The transformation of the sample had been on the surface, i.e. at most in the first μm . This surface nature of the nuclear transmutation has been almost a common characteristic observed by many researchers we have already notice several times (cf. e.g. [Kozima 1998 (Section 12.16), 2006 (Section 2.2.1.3), 2020 (Section 2.2)]).

“We have studied systems composed by several kinds of metal samples, such as pure Ni, nickel alloys and nickel plated, of a cylindrical or planar shape, in a hydrogen atmosphere. In a typical experiment the samples were inserted in a cell, loaded with hydrogen at pressure in the range of 100-1000 mbar and kept at temperatures between 420 and 720 K[1-3].” [Campari 2004a (Introduction)].

“The sample is a cylindrical rod made by a nickel alloy $\text{Ni}_{7.6}\text{Cr}_{20.6}\text{Fe}_{70.4}\text{Mn}_{1.4}$ with length 9.0 cm. A chemical and a physical cleaning are performed before of sealing the sample in the experimental cell, showed in Fig. 1 and described in Ref. 3. In the cell, annealing cycles are performed in vacuum and in a hydrogen atmosphere with the temperature in the range 400 to 700 K. The gas pressure is maintained in the range 100 to 1000 mbar.” [Campari 2004b (p. 414)]

“To correctly compare the components of the alloy, we note that the changes are essentially on the surface, i.e. at most in the first μm . This fact is confirmed by the elemental analysis performed in the central region showed in Fig. 5c, which is identical to an analysis of the blank.” [Campari 2004b (p. 417)]

4.1.7 Constantan Wires Coated by Sub-micrometric Surface Sponge [Celani 2012 – 2020b]

The extensive investigation of the constantan wire coated by sub-micrometric surface layer has been performed by Celani et al. [Celani 2012 – 2020b]. We will give rather minute introduction to their work in another paper presented in this Conference [Kozima 2021a (Sec. 2.3.2 and Appendix A5)].

We have given a brief explanation of the nuclear transmutation obtained in a multiply nano-coated Ni wire [Kozima 2011b].

The nuclear transmutation of several elements confirmed by the experiment is qualitatively explained assuming the single neutron absorption by elements in the surface layer. The decrease of Ru might be explained by absorption of a single n - p cluster.

4.1.8 Pd/Ni/Zr ($\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$) and Cu/Ni (CuNi_y) [Kitamura 2014 – 2018]

Kitamura et al. [Kitamura 2014 – 2018] had performed extensive investigations on the Pd-Ni and Cu-Ni nanocomposite CF materials, Pd/Ni/Zr ($\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$) or Cu/Ni (CuNi_y), supported by Zirconia (ZrO_2) or by Silica (SiO_2). These CF materials are composite and compound ones from our classification but have ambiguity about their structures and we take up them in this paper leaving the problem related to the interfaces to another paper.

We give a brief survey of the common characteristics of the works by Kitamura et al. in addition to a brief comment given recently [Kozima 2020] and the detailed introduction to the experimental data obtained by them will be given in Appendix A5.

Several of the characteristics obtained in the works by Kitamura et al. are listed as follows; (1) the excess heat is observed only in the elevated temperature phases of the runs with binary nanocomposite samples, but not with single-element nanoparticles. (2) Excess power from Pd-Ni nanocomposite CF materials at elevated temperature of 200 ~ 300 °C depend strongly on the Pd/Ni ratio, which will be one of the key factors to increase the excess power.

Considering the catalytic action of the electrodes noticed already long ago by the electrochemists [Bockris 1970a, 1970b, 2000, Horiuti 1970, Kita 1971] and the common knowledge listed in the general guidance to the catalysis (e.g. [Wikipedia Catalysis]), these characteristics cited above and others observed by Kitamura et al. show clearly that the CFP observed by them (AHE, the anomalous heat effect, in their terminology) is the nuclear reactions induced by the common mechanisms to the CFP observed hitherto in various CF materials summarized in several reviews (e.g. [Kozima 2019c]) and books (e.g. [Kozima 1998, 2006, Storms 2007]) characterized by the catalytic action of electrocatalysis [Bockris 1970a, 2000].

4.2 Ceramics

There are several works on the CFP in ceramics by Romodanov et al. [Romodanov 1998b] and Mizuno et al. [Mizuno 1996, 1997]. We give a brief introduction of the data in this section and will give an overview on the data obtained in ceramics and others by Romodanov et al [Romodanov 1993, 1998a, 1998b, 1998c] in Appendix A5.

There are several interesting investigations on the new ceramics possible to expect the CFP in them. A brief survey on these new ceramics is given in Appendix A4.

Romodanov et al. [Romodanov 1993, 1998a, 1998b, 1998c] had performed extensive

experiments with CF materials including transition metals, alloys (including SS) and ceramics (including TiC, VC, ZrB₂) in glow discharges with D₂, H₂ and D₂ + H₂ gases.

They observed generation of neutron, tritium and gamma radiation from cathode materials at elevated temperatures up to 1200 °C. They determined the maximum $N_n/N_t|_{\max}$ and minimum $N_n/N_t|_{\min}$ ratios of neutron and tritium generation in transition metal cathodes including Mo, Nb, Ta ranging $N_n/N_t|_{\max} = 8.5 \times 10^{-6}$ to 0.6×10^{-9} , $N_n/N_t|_{\min} = 1.7 \times 10^{-3}$ to 8×10^{-7} [Romodanov 1993].

In the case of ceramics cathode, they observed tritium generation depending on the species of the ceramics [Romodanov 1998b] a part of which had been explained in Sec. 4.1.2. Further explanation of the data by Romodanov et al. is given in Appendix A5.

Mizuno et al. observed excess heat and nuclear transmutations in the CF materials composed of Sr and Ce oxides (SrCeO₃) charged with D₂ gas at elevated temperatures ranging from 200 to 700 °C [Mizuno 1996, 1997].

“A coin-shaped proton conductor made from metal oxides of strontium and cerium can be charged in a hot D₂ gas atmosphere to produce excess heat. Anomalous heat evolution was observed from the proton conductors charged with alternating current at 5 to 45 V at temperatures ranging from 400 to 700 °C. The anomalous heat produced temperature increases as much as 50 °C. Excess heat was estimated as a few watts in most cases, totaling up 10 several kilojoules.” [Mizuno 1996 (Abstract)].

“Abstract-

A proton conductor, the solid state electrolyte, made from an oxide of strontium, cerium, niobium and yttrium can be charged in a hot D₂ gas atmosphere. Anomalous radioisotopes were detected in all samples charged with an alternating current with voltages ranging from 5 to 45 V, at temperatures ranging from 400 to 700 °C. No radioisotopes were detected from the sample treated in a H₂ gas atmosphere. The radioisotopes may be induced from a catalytic reaction between the metal and oxide interface to deuterium atoms.” [Mizuno 1997 (Abstract)].

4.3 Polymers; XLPE and Biological Systems

We have investigated already the CFP in XLPE [Kozima 2007, 2010, 2016a, 2019c] and biotransmutation in the glowing biological cultures [Kozima 1996, 2016b, 2019c] using the TNCF model to give a unified explanation of the observed results.

In biological systems, it should be mentioned a necessity of a microscopic

investigation including atomic processes using knowledge developed in electrochemistry (cf. e.g. [Woodbury 1970]) to clarify the mechanism working there for the CFP which will be given in another paper to be published recently [Kozima 2021c].

There are many ceramics which can contain a lot of hydrogen isotopes forming a regular lattice of host elements and hydrogen isotopes like those we met in XLPF and biological systems. We show several examples of them in Appendix A3 as future candidates where we can expect the CFP according to the mechanism working in XLPE and biological systems.

5 Conclusion

The necessary conditions to realize the CFP include two processes governed by the complexity:

- (1) *Adsorption of hydrogen isotope atoms (H/D), molecules(H_2/D_2) or ions (H^+/D^+)* by the host material composed of elements X's (A_ZX 's). Adsorption of ions on the electrode is a subtle problem of catalytic nature in the solid-liquid interface investigated extensively in electrochemistry (cf. e.g. [Bockris 1970a, 2000, Quaino 2007, Seo 2012]).
- (2) *Generation of the superlattice XH/XD (${}^A_ZX-p/{}^A_ZX-d$)* by the self-organization as a nonlinear process under a nonequilibrium condition belonging to the complexity. [Kozima 2013, 2014] (cf. also Section 1.1 of another paper presented in this Conference [Kozima 2021b]).

In the above two processes to fill the necessary conditions for the CFP in the composite CF material (details should be referred to the cited papers), we must take into our consideration the specific atomic processes in alloys and ceramics different from those in simple host elements like Ni and Pd.

In this paper, we have taken up experimental data sets on several features of the composite CF materials obtained from the early days to recent few years considering recent tendency to concentrate research works on them.

The experimental results obtained in the composite CF materials have shown the same characteristics as those observed in the CF materials with rather simple host materials. This fact suggests us that the cf-matter composed of the trapped neutrons and the neutron drops ${}^A_Z\Delta$ has a common nature and it is possible to investigate the CFP in the composite CF materials using the TNCF and ND models which have shown their usefulness to understand the CFP observed hitherto.

However, there are characteristics of the composite CF materials different from the CF materials composed of simple host materials. Since Claytor et al. [Claytor 1998] had shown twenty years ago, the composite CF materials have shown their ability to realize

the CFP until now as we pointed out in this and another paper which is presented in this Conference [Kozima 2021a]:

“Alloys have been discovered that gave much enhanced tritium output over randomly selected materials.” [Claytor 1998 (pp. 92, Conclusion)]

The characteristics of alloys and ceramics responsible to the aptitude for the CFP are listed up as follows from our point of view: (1) Elevated temperature of their operation up to 300 °C ($\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$) – 1000 °C (constantan) and more than 1500 °C in the case of ceramics (cf. [Kozima 2013 (Sec. 3.1), 2020]), (2) Distribution of minor elements at disordered sites (cf. [Kozima 2019b (Sec. 3-4)]), (3) Distribution of protons and deuterons at disordered sites (cf. [Kozima 2019b (Sec. 3-4)]), and (4) Interfaces around the CF material (cf. [Kozima 2021a]) which intensify the effects (2) and (3) in this list. The characteristic (4) is extensively discussed in another paper presented at this Conference [Kozima 2021a].

In the explanations of the CFP obtained in the composite CF materials, we want to emphasize the interesting facts observed by Fleischmann et al. [Fleischmann 1989] and by Claytor et al. [Claytor 1998] as explained in Sec. 2.4.1 and Sec. 4.1.3, respectively.

In these two cases, curious events related to both the formation of the cf-matter and the nuclear reactions between a trapped neutron and nuclei at disordered sites in the CF material have been observed and explained by our model.

The experimental data discussed in this paper have several characteristics depending on the several factors of the sample. One of the factors is the interfaces surrounding the CF material which is investigated in another paper presented in this Conference [Kozima 2021a]. Several of the factors not taken up at present due to the lack of space will be investigated in future.

We will give a justification of our approach to the composite CF materials using the TNCF model in another paper presented at this Conference [Kozima 2021b] where we use an analogy between neutron and electron energy bands.

Acknowledgements

The author would like to express his thanks to Dr. Y. Iwamura and Dr. A. Kitamura for the valuable communications on their experiments analyzed in Sec. 4.1.5 (Iwamura et al.) and Sec. 4.1.8 and Appendix A6 (Kitamura et al.). He is indebted also to Dr. T. Ohmori for valuable discussions on the hydrogen electrode reaction (HER) and the underpotential deposition (UPD).

Appendices

Appendix A1. History of the Acceptance of the TNCF Model

Appendix A2. Diffusivity of hydrogen isotopes in Mo, Nb and Ta [Voelkl 1978, Tanabe 1992] .

Appendix A3. Diffusivity of Hydrogen Isotopes in Ni, Pd and Cu [Voelkl 1978]

Appendix A4. Characteristics of Ceramics containing Hydrogen Isotopes [Kageyama 2020, Kobayashi 2020]

Appendix A5. Experiments at the Elevated Temperature by Romodanov et al. [Romodanov 1993, 1998a, 1998b, 1998c]

Appendix A6. Experiments with the $\text{Pd}_x\text{Ni}_{0.35-x}\text{Zr}_{0.65}$, CuNi_y , and others at the Elevated Temperature by Kitamura et al. [Kitamura 2014, 2018]

Appendix A1. History of the Acceptance of the TNCF Model

It is interesting to notice the history of the acceptance of a phenomenological approach as a possible trial to the science of the cold fusion phenomenon. There are several researchers who have shown their tolerance for the TNCF model in whom we can count John Dash, Makoto Okamoto and Koji Fusimi and some others who have been a personal acquaintance of the author needless to say the colleagues and co-authors of the present writer (H.K.). We cite some other people who recognized the value of the phenomenological approach in their course of investigation on the CFP.

A1-1 J.O'M. Bockris

It is interesting to notice that Prof. J.O'M. Bockris was only one in the CF researchers expressed explicitly an affirmative comment on the TNCF model showing his genuine scientific spirit and his correct understanding of the model. [Bockris 1999a, 1999b]

“Three points summarize the historical significance of the discovery of LENR:- - -

3 The work must give rise to modifications of the theory of the nucleus. However, new physics is probably not needed. Classical nuclear physicists have maintained an agnostic stance for so long because their knowledge of fusion is concerned with reactions in plasma, and they have paid little attention to the effects of a solid lattice upon nuclear activity within it after injection of H and D at high fugacity, or to the effect of free neutrons in the lattice.” [Bockris 1999a (p. 71)] (Underlines are added at citation)

“An all embracing theory must account for the long (500 hours) delay in a switch on of nuclear effects in electrolysis of D-Pd-LiOD, irreproducibility that is diminished by the

use of codeposition of D_2 and Pd on Au, and by use of thin films; internal damage in Pd; burst-like nature of the phenomena in Pd; occurrence in organic and biomaterials. No theory yet published accounts for all these effects. - - - Kozima⁵ has made a detailed case in which neutrons in the ambient enter all substances and are trapped there. These entities then undergo various reactions with Li, H, D, T, etc., and produce the observed effects. Although Kozima's model serves to explain the widespread nature of the effects (he sees neutrons as being "in" everything), his numerical analyses show the required neutron concentration to vary from 10^4 to 10^{13} cc^{-1} in D-Pd to achieve consistence, with results and this seems too large a range for consistency. Stress is put in Kozima's model on reaction with Li^6 but it was shown in 1990 by Appleby and Srinivasan²⁹ at Texas A&M that there was no difference in the heat evolved in Pd-LiOD in systems containing only Li^6 and those containing natural Li." [Bockris 1999b] (Underlined at citation)

[Kozima⁵] 5. Kozima, H. *Discovery of the Cold Fusion Phenomenon*, Ohtake Shuppan, Inc., Tokyo, Japan. [Kozima 1998]

[Srinivasan²⁹] 29. Appleby, A. J. and Srinivasan, S. 1990. *ICCF-1*, Salt Lake City.

In these sentences, Professor Bockris cited the trapped neutron as a probable new idea to explain seemingly contradicting experimental data such as the relations between numbers of tritium, neutron and helium-4 known already at that time. It is interesting to notice his comment on the choice of models in the *Preface* of his book published in 1970: "*They have tried to present, with due admission of the existing imperfections, a simple version of that model which seemed to them at the time of writing to reproduce the facts most consistently. They have for the most part refrained from presenting the detailed pros and cons of competing models in areas in which the theory is still quite mobile.*" [Bockris 1970a (p. vii)] (Underlined at citation)

A1-2 D.R.O. Morrison

Prof. D.R.O. Morrison, a well-known cons against the CFP from the early days of the history of the CF research, has shown his correct understanding of the TNCF model as his explanation of the lack of radiation in the nuclear reaction in the CFP.

[Morrison 2000]

"12. H. Kozima (abs. 044, 045, 046) Trapped Neutron Catalyzed Fusion, TNCF model. *Energy band of neutrons interacts coherently with lattice nuclei.*

e.g.

$n + {}_{46}^{100}\text{Pd} \text{ -----} > {}_{13}^{27}\text{Al} + {}_{33}^{67}\text{As}$ or $\text{-----} > {}_{26}^{56}\text{Fe} + {}_{20}^{44}\text{Ca}."$

[Morrison 2000 (Appendix 2, Theories at ICCF-8)] (italicized partially at citation).

In this citation, Morrison has shown his correct understanding of the TNCF model as his explanation of the lack of radiation in the nuclear reaction in the CFP as “*Energy band of neutrons interacts coherently with lattice nuclei.*” Almost ten years after the discovery of the CFP, Dr. Morrison as a scientist seemed to arrive at recognition that there is something reflecting truths behind the vast pile of experimental data and became more tolerant about assumptions to explain the facts inconsistent with the framework of the nuclear physics at that time.

A1-3 DOE Report 2004

In the DOE Report 2004 [DOE 2004], there are several positive evaluations for the works in the CFP. We cite only one of them related to the TNCF model.

[DOE 2004]

“(1) H. Kozima *J. Electroanalytical Chemistry* **425** 173 (1997), ISSN: 1572-6657.

Theory is presented using the ${}^6\text{Li}$ atom in the electrolyte of many experiments to provide a neutron that begins an interesting series of reactions leading to $d + d$ fusion.

(2) H. Kozima *J. Hydrogen Energy* **25** 505 (2000)

Use of his theory to understand Tritium production in others experiments.

(3) H. Kozima *J. Hydrogen Energy* **25** 509 (2000)

This article gives branching ratios:

$d + d = t$ (1.01 MeV) + p (3.02 MeV), (1)

$d + d = {}^3_2\text{He}$ (0.82 MeV) + n (2.45 MeV), (1)

$d + d = {}^4_2\text{He}$ (76 keV) + γ (23.8 MeV), (10^{-6}).”

[DOE 2004 (Summary, Reviewer #4, pp. 7 – 8)] (Italicized partially at citation)

The reviewer #4 has cited three papers from ours published before 2004 as possible candidates for a unified explanation of the contradicting experimental data on the tritium, neutron and helium-4 and also of data obtained not only in deuterium but also in protium systems.

A1-4 E. Storms

On the other hands, Dr. E. Storms had shown that he did not understand the nature of the phenomenological approach at all.

[Storms 2007]

“Kozima^{180 – 183} at Shizuoka University (Japan) and Portland State University (US) proposes that solids contain clusters of neutrons. According to his TNCF (Trapped Neutron Catalyzed Fusion) theory, these are stabilized by a unique configuration and are

released to cause cold fusion reactions under certain conditions. Two of the many questions not answered are: ‘*Why is the extra mass added by these neutron structures not observed in normal material and why are neutrons not released in a detectable form when the stabilizing solid structure is vaporized?*’ ” [Storms 2007 (p. 135)] (Italicized partly at citation. The references [180] – [183] are those of the references in the original book [Storms 2007])

The “*Two of the many questions not answered*” raised by Storms in this sentence show that he does not understand the meaning of the phenomenological approach at all, to our regret.

A1-5 Conclusion

It should be mentioned again that there are almost no favorable mentions on the TNCF model in the cold fusion community despite a few supporters such as John Dash, Makoto Okamoto, and Koji Husimi needless to say colleagues and co-authors of the present author (H.K.). Only one exception is Prof. J.O’M. Bockris who had shown his genuine scientific spirit and his correct understanding of the model.

In this meaning, there have been a large contrast between true scientists, such open minded people looking for the truth behind the complicated experimental facts as pointed out above, and the “*Voodoo Scientists*” relying only on the second-hand articles (written in such books as [Huizenga 1992, Taubes 1993]) discarding and neglecting experimental facts in “the Medawar zone” [Kozima 1998 (Sec. 16.5, “A Message from the Right Side of the Medawar Zone” by Peter Gluck)]. It is a sad history in modern science to see many people remain as Voodoo Scientists even after about 30 years of piling up of vast positive facts and of sincere theoretical efforts.

In the Voodoo Scientist, there should be Dr. Bob Park who had criticized the CF research enthusiastically for many years not depending on the facts and had written many negative comments on the CFP and finally a book “Voodoo Science” [Park 2000] in which he used the word *voodoo* suggesting the meaning of that word used to define the “Voodoo Scientist” given above.

It is interesting to cite the definition of the Voodoo Science from Bob Park’s book:

*“The line between foolishness and fraud is thin. Because it is not always easy to tell when that line is crossed, I use the term **voodoo science** to cover them all: pathological science, junk science, pseudoscience, and fraudulent science. This book is meant to help*

the reader to recognize voodoo science and to understand the forces that seem to conspire to keep it alive.” [Park 2000 (p. 10)] (Italicized and colored at citation).

In relation to the above definition of the Voodoo Science by Bob Park, I would like to define the Voodoo Scientist as follows:

“A voodoo scientist is such a pseudoscientist who did not use experimental data to judge whether a science is a real science or is a pseudoscience as Bob Park himself as we see in his book ‘*Voodoo Science*’.”

It is surprising to read *at present* an article appeared in *Physics Today* (Feb. 17, 2021) by Harry Collins, a sociologist of science (by Wikipedia), ignorant of the today’s physics changed drastically in the last half of 20th century by the inclusion of complexity in its.

He said,

“In an area of serious dispute, you can’t tell who is right simply by repeating the experiments because scientists disagree about when replication has been done properly. So, in cases like this, scientists invent theoretical reasons for why it must be this way or it must be that way. They will argue, and eventually settle by broad agreement, that such an apparatus should or shouldn’t see gravity waves. It’s more like a social agreement than a transfer of information or a formula.

For example, to debunk Weber’s detection claims, scientists had to get others to agree that Weber made mistakes in his statistical analysis and his handling of the data. And to debunk cold fusion, it had to be agreed that [Stanley] Pons and [Martin] Fleischmann were not the right kind of scientists to be doing the work. In neither case was it enough, at the time, simply to say the results weren’t replicated, even though that is how we describe it in retrospect. In 2015, with the first accepted discovery of gravitational waves, that kind of accusation could not be made to stick, even though some people tried it.” (Italicized and colored partially at citation.)

Harry Collins do not entirely understand that the quantitative reproducibility is a specific attribute of the physics governed by the linear dynamics and therefore we must satisfy with the qualitative reproducibility discarding the quantitative one in the physics like the cold fusion phenomenon where events are governed by the nonlinear interaction between particles in the system [Kozima 2013, 2019b].

It is really a very difficult work to do science rightly avoiding many bypaths leading to mistaken results. In the history of the cold fusion research, the most important characteristics in this science may be the complexity of the cold fusion phenomenon destined by the nonlinear interaction between participating particles [Kozima 2013] and

the meta-analysis of the data handling due to the characteristics of the CF materials [Kozima 2019a]. If we forget this fact, we will lose the way.

Appendix A2. Diffusivity of hydrogen isotopes in Mo, Nb and Ta. [Voelkl 1978, Tanabe 1992]

Romodanov et al. had measured tritium production in many CF materials including various transition metals Y, Mo, Nb, Er, Ta, and W and ceramics TiC, VC, ZrB, ZrC, ZrN, and LaB at high temperature realized by glow discharges and electric heaters (Appendix A4). We can contemplate the cause of the CFP in those transition metals at higher temperatures up to 1000 – 2000 K considering the diffusivity of hydrogen isotopes in Mo, Nb, Ta and Pd (for comparison) as shown in Figs. A2-1 – A2-4.

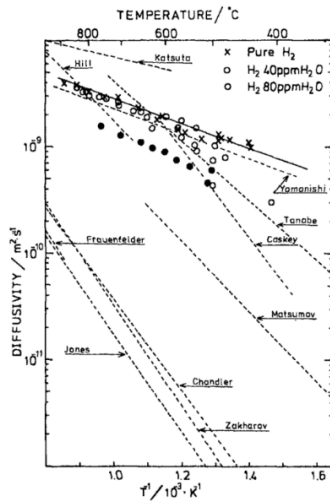


Fig. A2-1. Temperature dependence of the diffusivity for pure H₂, H₂ with 40 and 80 ppm H₂O in Mo. Literature data [8,9,11,16- 22] are also shown for comparison [Tanabe 1992].

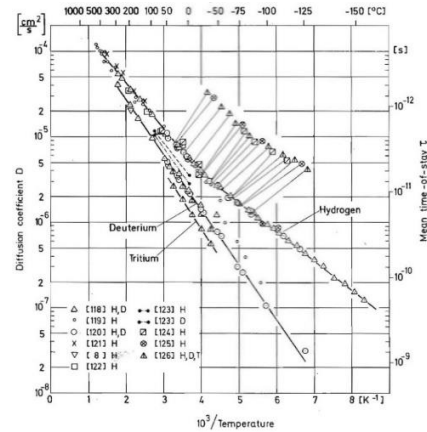


Fig. A2-2. Diffusion coefficient of H, D, and T in Nb. The mean time-of-stay τ is calculated for tetrahedral-tetrahedral jumps. (Numbers in brackets refer to References cited in [Voelkl 1978 (Fig. 12.5)].

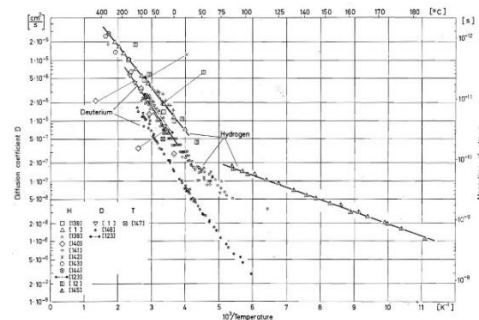


Fig. A2-3. Diffusion coefficient of H, D, and T in Ta.

and T in Ta. In addition, results with Pd-coated samples [12.123] are shown. Nevertheless, discrepancies in the absolute value (not so much in the slopes) can be note. (Numbers in brackets refer to References cited in [Voelkl 1978]) [Voelkl 1978 (Fig. 12.6)].

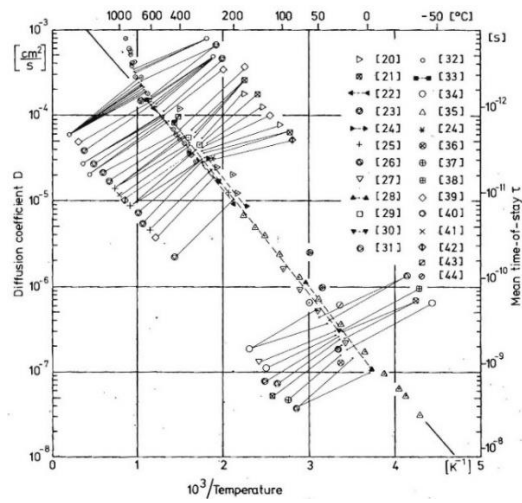


Fig. A2-4. Diffusion coefficients of H in Pd. The right-hand scale for the mean time-of-stay τ refers to octahedral-octahedral jumps. (Numbers in brackets refer to the paper cite in [Voelkl 1978]) [Voelkl 1978 (Fig. 12.2)].

Compare the value of the diffusion coefficients D (cm²/s) in Mo, Nb and Ta with that in Pd, $D = 2 \times 10^{-5}$ cm²/s in Pd at 200 C: $D = 1 \times 10^{-9}$ cm²/s in Mo at 400 C, 5×10^{-5} cm²/s in Nb at 200 C, and 2×10^{-5} cm²/s in Ta at 200 C. We can see the necessity of high temperature for Mo to realize the CFP compared with Pd which have been used very often in the experiments in the CFP. The durability of Pd over Nb and Ta is advantageous for the experiments and the latter two have not used often hitherto.

Appendix A3. Diffusivity of Hydrogen Isotopes in Pd, Ni and Cu [Voelkl 1978]

As the data shown below (Fig. A3-1 – A3-4), the diffusion coefficients of hydrogen isotopes in Pd, Ni, and Cu have similar values in the elevated temperature region above 800 °C. It is interesting to notice that they have almost the same values in Ni and Cu at around the room temperature (300 °C).

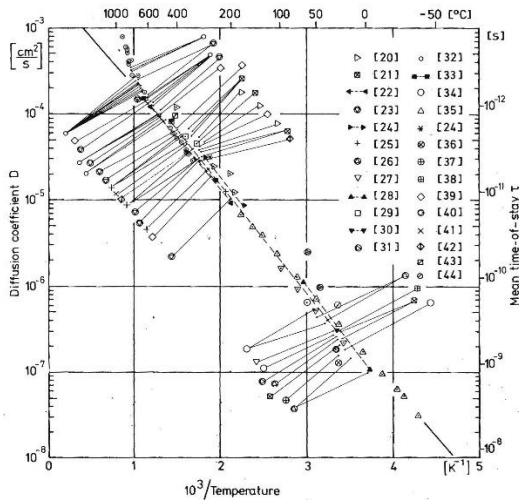


Fig. A3-1. Diffusion coefficients of H in Pd. The right-hand scale for the mean time-of-stay τ refers to octahedral-octahedral jumps. (Numbers in brackets refer to References of the original paper) [Voelkl 1978 (Fig. 12.2)]

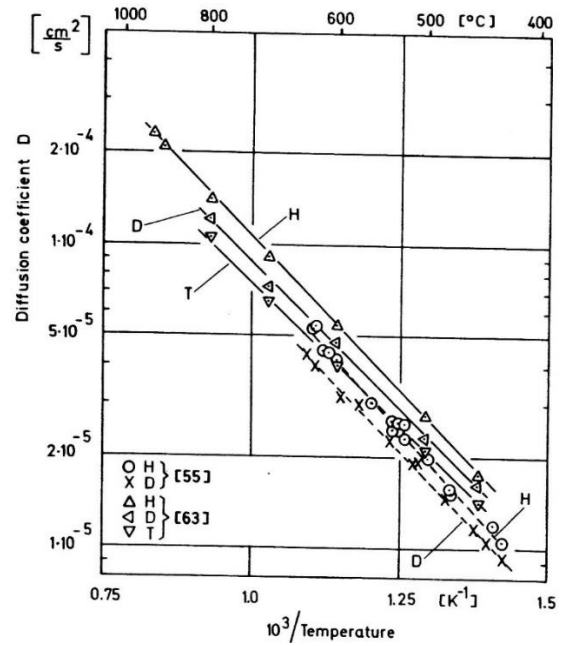


Fig. A3-3. Diffusion coefficient of isotopes of hydrogen in Cu (Numbers in brackets refer to References in the original paper) [Voelkl 1978 (Fig. 12.18)].

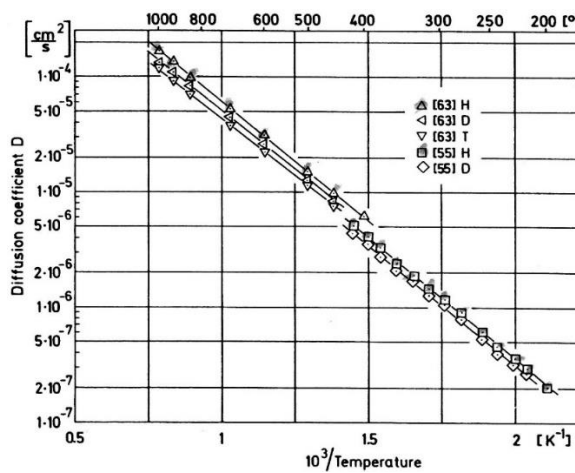


Fig. A3-2. Diffusion coefficient of isotopes of hydrogen in Ni (Numbers in brackets refer to References) [Voelkl 1978 (Fig. 12.17)].

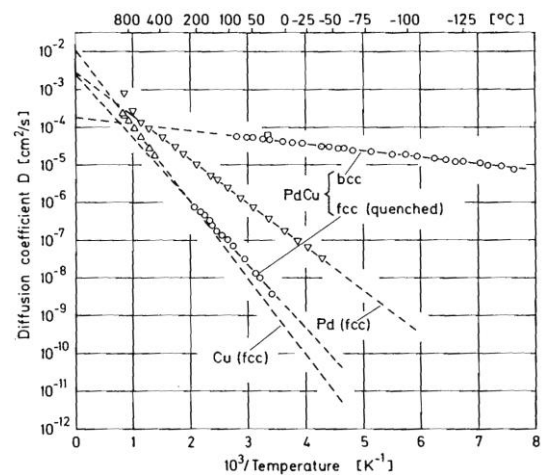


Fig. A3-4. Diffusion coefficient D for Pd, Cu [12.63] and $\text{Pd}_{0.47}\text{Cu}_{0.53}$ (bcc) [12.179] and $\text{Pd}_{0.47}\text{Cu}_{0.53}$ (fcc) [12.180]; \square = Piper [12.174] [Voelkl 1978, (Fig. 12.21, numbers in square bracket $[\]$ are references in the original paper) [Voelkl 1978 (Fig. 12.21)]

Appendix A4. Crystal Structures and Characteristics of Ceramics; TiC, VC, ZrB₂, ZrC, ZrN, LaB₆ [Kageyama 2020, Kobayashi 2020]

There are several CF experiments on the ceramics such as by Romodanov et al. [Romodanov 1998b] and Mizuno et al. [Mizuno 1996, 1997a]. We want to point out further possibility to observe the CFP in ceramics which have not observed it until now.

There are a few ceramics which can contain a lot of hydrogen isotopes as shown by many researchers (e.g. [Kageyama 2020, Kobayashi 2020]). Considering our knowledge about the CFP in XLPE [Kozima 2016a, 2019c] and biological systems [Kozima 2016b, 2019c], we must pay attention to them for the CF materials even if they did not show the nuclear reactions in them yet. We show some examples of their structure in Figs. A3-1 – A3-4 [Kageyama 2020, Kobayashi 2020] which remind us the structure we know in the XLPE and biological systems where the CFP had been observed.

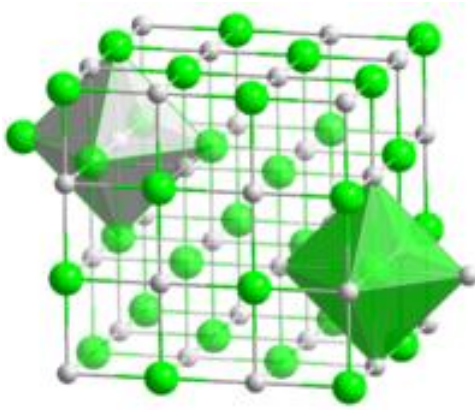


Fig. A4-1. Zirconium nitride ZrN

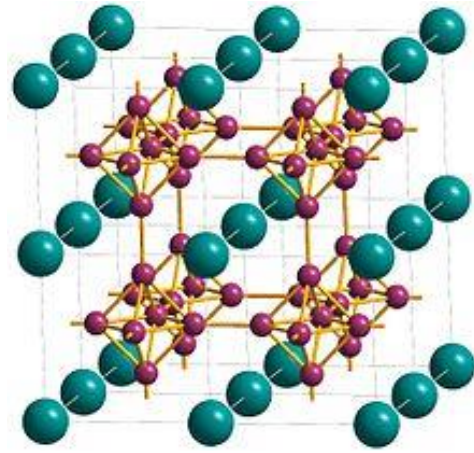


Fig. A4-3. Lanthanum hexaboride LaB₆

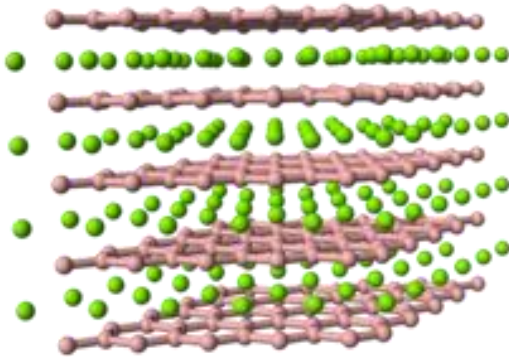


Fig. A4-2. Zirconium Diboride ZrB₂

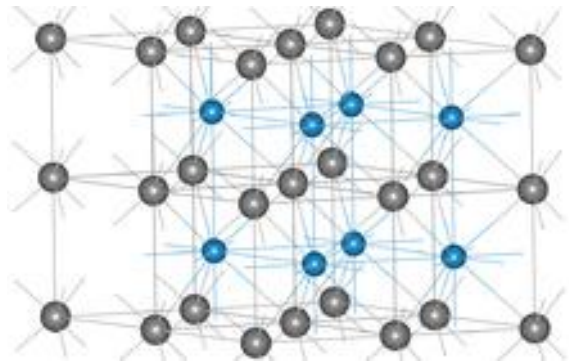
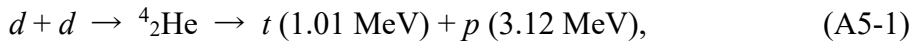


Fig. A4-4. α -WC structure, carbon atoms are gray.

Appendix A5. Experiments at the Elevated Temperature by Romodanov et al. [Romodanov 1993, 1998a, 1998b, 1998c]

In the three papers by Romodanov et al. [Romodanov 1993, 1998a, 1998b, 1998c], they observed tritium from three kinds of CF materials; (1) Transition metals Y, Mo, Nb, Er, Ta, and W [Romodanov 1993], (2) H/D transfusion through V, Nb, Ta and Mo, (3) ceramics TiC, VC, ZrB₂, ZrC, ZrN, and LaB₆ [Romodanov 1998b], and (4) transition metals Fe, Ti, Nb and an alloy Fe-Cr-Ni-Ti (71-18-10-1) [Romodanov 1998c]. In the cases (1) (2) and (3), the glow discharges with large currents were used to heat the samples, and in the case (4) electric heater was used instead.

It is apparent that the purpose of the experiments (1) and (2) is detection of the nuclear products by the $d-d$ fusion reactions in these CF materials using the accelerated deuteron onto the targets where are occluded enough deuterium. However, the acceleration of deuteron by the glow discharge was insufficient to realize the expected nuclear reactions.



We have to understand the effect of the glow discharge in these experiments (1) – (2) is to heat these CF materials up to around 1000 K and to feed deuterium into them which are difficult to absorb and occlude hydrogen isotopes in lower temperatures than 300 K. In all experiments performed at the elevated temperature more than 1000 K, sometimes up to 2100 K, they observed generation of tritium, an event characteristic of the CFP. These experiments show clearly the elevated temperature is favorable for the CFP. The large amount of the observed tritium is explained by the reaction (A4) written down below in consistent with other various events in the CFP as shown in our works [3, 4, 5]:



where φ 's expresses phonons excited by the freed excess energy of ${}^3_1\text{H}^*$ [3, 4] replacing the gamma in the reaction in free space written down as follows:



The necessity of high temperatures for the Mo (and for ceramics) is discussed in Appendix A1 using the data of diffusivity in Mo compared to that in Pd. Unfortunately, we have no data of diffusivity in ceramics which may be comparable to or smaller than that of Mo at around room temperature.

“Table 2

Ratio of neutron and tritium fluxes when irradiating a number of materials by deuterium ions.” [Romodanov 1993 (p. 313, Table 2)]

Table 2

Ratio of neutron and tritium fluxes when irradiating a number of materials by deuterium ions

Material	Process parameters			Background, pulse 100 s	Reduced activity in view of background, Bc/ml	Ratio of deuterium activities after and before experiments	Tritium atom flux, atom/s	Nuclear interaction coefficient, atom/ion	Neutron-to-tritium flux ratio	
	E, eV	T, K	τ , h						min	max
D ₂	-	-	-	240	130	-	-	-	-	-
Y	40	1170	80	210	290	2.2	$1.2 \cdot 10^5$	$4.1 \cdot 10^{-15}$	$8.5 \cdot 10^{-6}$	$1.7 \cdot 10^{-3}$
Y	80	1270	23	250	$1.2 \cdot 10^3$	9.2	$4.5 \cdot 10^6$	$1.6 \cdot 10^{-12}$	$2.2 \cdot 10^{-7}$	$4.4 \cdot 10^{-5}$
Mo	125	1470	10	150	$6 \cdot 10^3$	46	$4.5 \cdot 10^6$	$9.2 \cdot 10^{-12}$	$2.2 \cdot 10^{-7}$	$4.4 \cdot 10^{-5}$
Mo	100	970	10	170	$1.6 \cdot 10^3$	12.3	$1.8 \cdot 10^7$	$5.9 \cdot 10^{-12}$	$5.5 \cdot 10^{-8}$	$1.1 \cdot 10^{-5}$
Nb	75	1170	162	230	$4.7 \cdot 10^4$	$3.6 \cdot 10^2$	10^7	$3.8 \cdot 10^{-13}$	10^{-7}	$2 \cdot 10^{-5}$
Nb	80	1170	60	700	$3 \cdot 10^6$	$2.3 \cdot 10^4$	$1.7 \cdot 10^9$	$6.8 \cdot 10^{-11}$	$0.6 \cdot 10^{-9}$	$1.8 \cdot 10^{-7}$
Nb	100	1670	8	240	$5.5 \cdot 10^4$	$4.2 \cdot 10^2$	$0.9 \cdot 10^9$	$1.2 \cdot 10^{-10}$	$1.1 \cdot 10^{-9}$	$2.2 \cdot 10^{-7}$
Er	50	1070	140	460	$1.2 \cdot 10^3$	9.6	$3.1 \cdot 10^5$	$9.9 \cdot 10^{-15}$	$3.2 \cdot 10^{-6}$	$6.4 \cdot 10^{-4}$
Er	70	1270	6	530	$8.9 \cdot 10^2$	6.8	$1.5 \cdot 10^7$	$3.2 \cdot 10^{-12}$	$6.7 \cdot 10^{-8}$	$1.3 \cdot 10^{-5}$
Ta	70	1570	110	350	$3.1 \cdot 10^3$	23.5	$9.6 \cdot 10^5$	$3.6 \cdot 10^{-14}$	$1.1 \cdot 10^{-6}$	$2.1 \cdot 10^{-4}$
Ta	90	1670	5	180	$1.6 \cdot 10^3$	12.3	$3.4 \cdot 10^7$	$7.3 \cdot 10^{-12}$	$2.9 \cdot 10^{-8}$	$5.9 \cdot 10^{-6}$
W	70	1500	115	600	$8.5 \cdot 10^5$	$6.5 \cdot 10^3$	$2.5 \cdot 10^8$	$9.1 \cdot 10^{-12}$	$4 \cdot 10^{-9}$	$8 \cdot 10^{-7}$
W	110	1670	10	760	$1.3 \cdot 10^4$	10^2	$1.7 \cdot 10^8$	$2.5 \cdot 10^{-11}$	$5.8 \cdot 10^{-9}$	$1.2 \cdot 10^{-6}$

“The tritium generation rate has increased by four orders of magnitude, while increasing the specific power by a factor of four; and it has reached the value of 10^9 atom/s when the neutron-to-tritium yield ratio is in the range from 10^{-7} to 10^{-9} .” [Romodanov 1993 (p. 307)]

The data telling that the neutron-to-tritium yield ratio is in the range from 10^{-7} to 10^{-9} is consistent with the theoretical prediction of $N_n/N_t \approx 10^{-7}$ by the TNCF model cited in Eq. (1.1).

“- - deuterium transfusion through sample in plasma for V, Nb, Ta has not rendered the appreciable influence on tritium generation rate. For samples from molybdenum single crystal and polycrystal the deuterium transfusion resulted in increase of tritium generation rate in two times, in comparison with stationary conditions.” [Romodanov 1998a (p. 585)]

“It is shown, that the heaviest rate of tritium generation, of materials TiC, VC, ZrB₂, ZrC, ZrN, and LaB₆, was observed at bombardment by deuterium ions from plasma of glow discharge for ZrB₂ and made $3.6 \times 10^7 - 3.9 \times 10^7$ atoms/s, at efficiency $2.5 \times 10^{-12} - 2.9 \times 10^{-12}$ (? Exponents -12 is ambiguous) atom/ion, that is comparable with results for metal targets.” [Romodanov 1998b (p. 590)]

Translation of the sentence in the above paragraph.

“- - - At bombardment of materials TiC, VC, ZrB₂, ZrC, ZrN, and LaB₆ by deuterium ions from plasma of glow discharge, the heaviest rate of tritium generation was observed for ZrB.”

“4. Conclusion

4.1 The significant tritium generation rate, is fixed at radiation by ions deuterium of samples from borides, carbides and nitrides of titanium, vanadium, zirconium, and lanthanum from plasma powerful of glow discharge. For samples of ZrB₂, the tritium generation rate reached $(3.6 - 3.9) \times 10^7$ atom/s.

- - -

4.3 The most proof of materials, at radiation in plasma of glow discharge, were ZrB₂ and ZrN, and the least proof samples of LaB₆.” [Romodanov 1998b]

“The research is conducted on samples of iron, and its alloys as well as on samples of titanium and niobium at interaction, basically, with ordinary hydrogen. It is established, hydrogen that the short-term bursts of tritium generation, arising at interaction of isotopes in which is immersed, previously heated up metal sample, relate to output tritium, early activated, owing of effects isotopes.” [Romodanov 1998c (Abstract)]

Appendix A6. Experiments with Pd_xNi_{0.35-x}Zr_{0.65}, CuNi_y, and others at the Elevated Temperature by Kitamura et al. [Kitamura 2014 – 2018]

Kitamura et al. have developed the so-called “nano-composite samples of Pd-Ni alloys” on some stable solid materials such as silica (SiO₂) and zirconia (ZrO₂) and have worked out elaborate experiments on the large amount of excess heat when the materials were in contact with protium or deuterium gas in rather higher temperature above about 200 °C [Kitamura 2014, 2015, 2016, 2017, 2018]. It should be mentioned that their work was stimulated by the work developed by Celani et al. since 2011 as introduced in the Appendix 4 in another paper presented at this Conference [Kozima 2021a].

We review works by Kitamura et al. below according to the material used in the experiments.

We can characterize the experimental systems taken up by Kitamura et al. by two properties; (1) The composite CF materials and (2) the solid-solid interface between the CF material and the supporter. In this paper, we will concentrate our investigation on the first characteristic leaving the second to another papers [Kozima 2021a, 2021b, 2021c]. The first characteristic has close relation to the *supported catalysis* explained in Introduction of this paper [Wikipedia Catalysis].

A6-1. Silica based samples [Kitamura 2016, 2018]

“Abstract

Hydrogen isotope absorption characteristics of nanoparticles supported by silica, Pd/SiO₂ (“PSf1”) and Cu₁Ni₁₀/SiO₂ (“CNS3”), have been examined. Large absorption energy (1.3 ± 0.3 eV/Pd) with large apparent loading ratio (LM = 2.6) was observed in the initial phase of the D-PSf1#1 run with D₂ at room temperature, which could be ascribed to reduction of PdO and hydrogen absorption by Pd nanoparticles. To reduce the NiO in the CNS3 sample, heating up to around 200 °C was necessary. The excess heat was observed in the elevated temperature phases of the runs with CNS3, while no excess heat was observed with PSf1. Taking also into account the experimental results obtained previously for other samples, we can conclude that the excess heat is observed only in the elevated temperature phases of the runs with binary nanocomposite samples, but not with single-element nanoparticles. In the HCNS3#2 run, the excess heat amounts to 200 eV/Ni or more than 0.9 keV/H without detectable dose rate of hard radiations, which cannot be explained by any chemical process.” [Kitamura 2016]

A6-2. Zirconia based samples [Kitamura 2015, 2017, 2018]

“Abstract

Hydrogen isotope absorption characteristics of Ni-based nano-composites supported by zirconia, Pd_{0.044}Ni_{0.31}/ZrO₂ (“PNZt”) and Cu_{0.044}Ni_{0.31}/ZrO₂ (“CNZt”), have been examined. Large absorption/adsorption (sorption) energy of $(1.6 \pm 0.2) \times 10^1$ eV/atom-Pd or (2.3 ± 0.9) eV/atom-D in the initial phase of the #1 run with D₂ at room temperature was observed. If Ni is taken into account as the absorbent element, a rather plausible value of 2.0 ± 0.3 eV/atom-M (M stands for both Pd and Ni), yet one-order-of-magnitude larger

than the conventional absorption energy of 0.2 eV/H, is obtained. This means that Pd atoms act as a catalyzer for deuterium absorption of Ni. The large sorption energy was repeatable with about $(6\sim9)\times 10^{-1}$ eV/atom-M in D-PNZt#2 through H-PNZt#4 runs. In the elevated temperature (200 ~ 300 °C) phases, on the other hand, excess temperature of 15 ~ 16 °C corresponding to conservatively evaluated excess power of 11 ~ 12 W (or 1.3 ~ 1.5 W/g-Ni) was recorded repeatedly in both PNZt and CNZt sample runs with both H and D.” [Kitamura 2015 (p. 1 (Abstract))]

$\text{Pd}_{0.044}\text{Ni}_{0.31}/\text{ZrO}_2$ (“PNZt”) and $\text{Cu}_{0.044}\text{Ni}_{0.31}/\text{ZrO}_2$ (“CNZt”), have been examined. - - This means that Pd atoms act as a catalyzer for deuterium absorption of Ni. This is a typical case of the *supporter* for the catalysis [Wikipedia Catalysis].

“Abstract

Anomalous heat effect by interaction of hydrogen isotope gas and nanoparticles supported by zirconia, $\text{Pd}_1\text{Ni}_{10}/\text{ZrO}_2$ (“PNZ6” and “PNZ6r”) and $\text{Pd}_1\text{Ni}_7/\text{ZrO}_2$ (“PNZ7k”), has been examined. Excess power of 3 ~ 24 W from PNZ6 at elevated temperature of 200 ~ 300 °C continued for several weeks. The PNZ6 and PNZ6r samples with Pd/Ni=1/10 generated much higher excess power than PNZ7k with Pd/Ni=1/7. The Pd/Ni ratio is one of the key factors to increase the excess power. The maximum phase-averaged sorption energy, $\eta_{\text{av},j}$, exceeded 270 keV/D, and the integrated excess energy, E_a , reached 1 keV/Pd·Ni. It is impossible to attribute the excess energy to any chemical reaction; it is possibly due to some unidentified radiation-free nuclear process.

Index Terms – Zirconia supported nanoparticles, Pd·Ni/ZrO₂, hydrogen isotope gas, absorption and desorption, excess power, sorption energy of 270 keV/D. I” [Kitamura 2017 (p. 14, Abstract)]

Anomalous heat effect by interaction of hydrogen isotope gas and nanoparticles supported by zirconia, $\text{Pd}_1\text{Ni}_{10}/\text{ZrO}_2$ (“PNZ6” and “PNZ6r”) and $\text{Pd}_1\text{Ni}_7/\text{ZrO}_2$ (“PNZ7k”), has been examined. Excess power of 3 ~ 24 W from PNZ6 at elevated temperature of 200 ~ 300 °C continued for several weeks. The PNZ6 and PNZ6r samples with Pd/Ni=1/10 generated much higher excess power than PNZ7k with Pd/Ni=1/7. The Pd-Ni alloy shows a typical example of the *supported catalysis* [Wikipedia Catalysis].

A6-3. Review of the work from the viewpoint of the TNCF model [Kozima 2020]

The data finally compiled in the paper published in International J. Hydrogen Energy [Kitamura 2018] was briefly reviewed by us [Kozima 2020] and the essential result is

cited below.

It should be noted that the occurrence of the exothermic reactions accompanied with the crystallization of component elements in Pd-Ni alloys well-known in metallurgy of Pd alloys (cf. [Mizumoto 1989]).

“In the electroless plating of Pd-Ni-P alloys from ethylenediamine complex solutions containing sodium hypophosphite as a reducing agent, the generation of heat due to the crystallizations of palladium and palladium phosphides were observed at around 300°C. Another generation of heat was observed also at around 400°C due to the crystallization of Ni₃P₁. We found several broad peaks of the heat generation between the two peaks of the heat generations due to the Pd-P deposit and Ni-P deposit. Figure 5 shows the relation between the phosphor content in the deposit of Pd-Ni-P plating and amount of heat generation (determined by the peak of heat generation in the D_{sc} curve).” [Mizumoto 1989 (p. 68, Translated into English by H.K. The Fig. 5 in the above sentence is cited below as Fig. A6-1)]

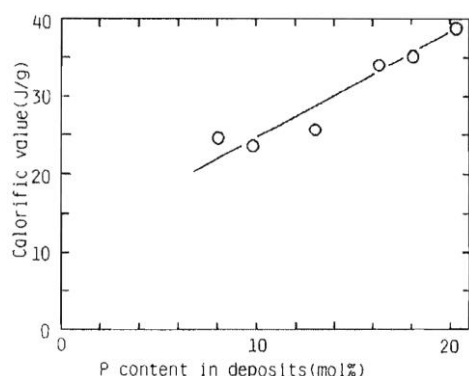


Fig. A6-1. Relation between P content in electroless Pd-Ni-P films and calories of exothermic reaction. ([Mizumoto 1989 (Fig. 5)])

It is probable to occur similar exothermic reaction in the hydrides/deuterides used in the experiments reported in the paper [Kitamura 2018]. Then, it is possible that there occurs a mutual acceleration of the positive feedback of the energy to the nuclear reactions in the CFP [Kozima 2013] and of the energy to the exothermic crystallization of component elements. The exothermic crystallization depends definitively on the composition of the CF material and therefore the process should be very complicated. If this mutual acceleration occurs, the CFP in the CF materials composed of Pd-Ni alloys works very effective to realize nuclear reactions between neutrons and nuclei at disordered position and then the structure of the CF material deteriorates rapidly to terminate the reactions by destruction of the optimum structure for the CFP [Kozima

2013]. So, it is desirable to check the occurrence of the crystallization in the sample after the experiments.

Acknowledgement

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