The 21st Meeting of Japan CF-Research Society

JCF21 ABSTRACTS

December 11-12, 2020

(Online Meeting)

Japan CF-Research Society

Evidence for Surface Heat Release Reaction over Nano-sized Multilayer Metal Composite with Hydrogen Gas

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We have been investigating the excess energy generation using a nano-sized multilayer metal composite with hydrogen gas [1]. Two nano-sized metal multilayer composite samples, which were composed of Ni, Cu, CaO thin films on bulk Ni (25mm×25mm×0.1mm), were used. These samples were fabricated by Ar ion beam or magnetron sputtering method. Heat burst and excess energy generation were observed during the experiments using nano-sized metal multilayer composite on Ni substrate and hydrogen gas. Heat burst phenomena were simultaneously detected by a radiation thermometer looking at the surface of the multilayer thin film and a thermocouple located near the metal composite. Maximum released excess energy reached 1.1MJ and average released energy per absorbed total hydrogen was 16 keV/H or 1.5 GJ/H-mol [1]. It cannot be explained by any known chemical process and suggests that the observed heat generation must be of nuclear origin. However, the reaction mechanism and the conditions to cause these phenomena to have not been clarified.

Recently, we have introduced additional radiation thermometers and surface temperature measurement for the two nano-sized metal multilayer composite samples became possible. Figure 1 shows temperature time variations for Tc, Surface A and B. It is possible to see that heat release reaction occurred at the surface A at first and afterwards at the surface B. Their heat bursts propagated to the Tc. This example gives us clear evidence that heat release reactions occur in the near surface region of the nano-sized multilayer metal composite with hydrogen gas.

Various analysis methods, such as SEM-EDX or TOF-SIMS, had been applied to obtain information about what kind of reactions are induced by the interaction of the nano-sized multilayer metal composite with hydrogen gas. Evidence for surface reaction will be also presented by the elemental analysis.

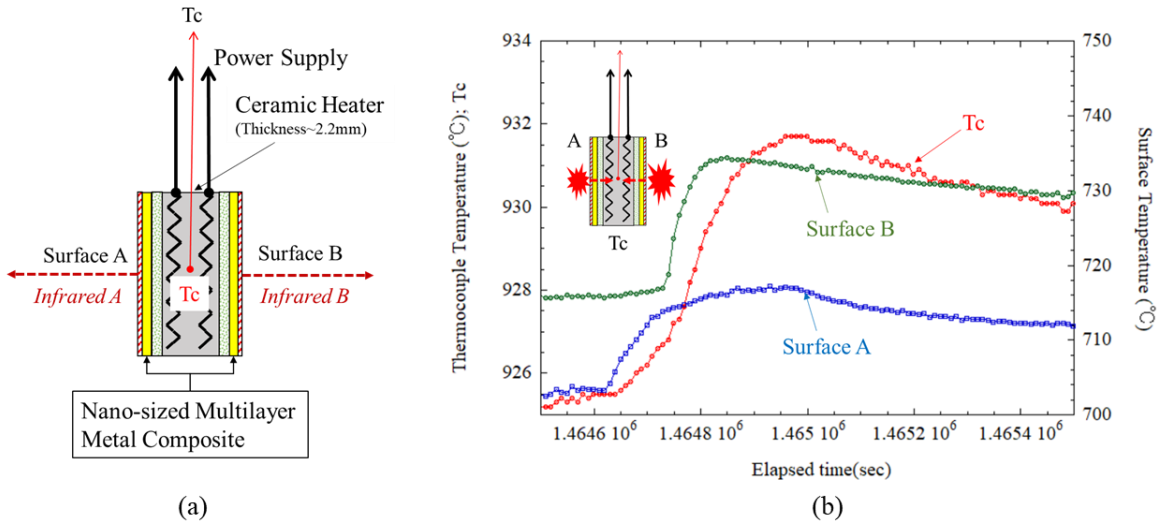

Figure 1. Evidence for surface heat release reaction; (a) Measurement points of Tc and Surface Temperature A and B, (b) Heat release reaction occurred at the surface A at first and afterwards at the surface B. Their heat bursts propagated to the Tc.

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H. Kozima JCF21, **21-1**,

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

Hideo Kozima*

Abstract

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) based on the phenomenological approach. In our phenomenological approach, we have used 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. The same approach is applied to deduce a consistent explanation of the specific experimental data obtained in the composite CF materials with rather complex host materials of various compositions and structures. We take up following CF materials in this paper: (1) alloys, (2) ceramics, and (3) polymers including XLPE and biological systems. The characteristics of the CFP observed in these composite CF materials are explained successfully by our model as we have done for the various experimental data sets 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% D_2O + 0.5% H_2O 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 $Fe_{71}Cr_{18}Ni_{10}Ti_1$ 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 $Cu_{55}Ni_{44}Mn_1$ [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 are explained from our point of view as briefly summarized in the beginning of this abstract.

The fundamental problems related to the premises of the model in relation to the composite CF materials will be developed in another paper presented in this Conference.

H. Kozima JCF21, **21-2**,.

Cold Fusion Phenomenon in the Compound CF Materials – Effects of Interfaces –

Hideo Kozima*

Abstract

In our phenomenological approach, the necessary and sufficient condition for the cold fusion phenomenon (CFP) has been established as the formation of the neutron energy bands in the super-lattice of host elements and the hydrogen isotopes.

In this paper, we take up the cold fusion phenomenon (CFP) observed in the compound (combined) CF materials, i.e. CF materials composed of host elements (Pd, Ni, C, etc.) and hydrogen isotopes (H and/or D) with interfaces between the environment or the substrates..

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 compound CF materials with various structures.

We take up following CF materials in this paper: **(1)** CF materials with interfaces between liquids and gases surrounding them, **(2)** CF materials with substrates, **(3)** Multi-layered CF materials. The characteristics and specific features of the CFP observed in these compound CF materials are explained successfully by our model.

Several examples of the events observed in compound CF materials taken up in this paper are; **(a)** specific (localized) sites in the near surface region (of deuterated Ti samples) resulting in 10^{10} to 10^{12} fusion reactions during each event [Srinivasan 1990a], **(b)** the Pd/D codeposition. (on Cu substrates) [Szpak 1991a], **(c)** tritium and neutron results that have been obtained with the Pd-Si-D system [Claytor 1991b], **(d)** analyses of used cathodes have revealed the presence of several light elements in the near-surface region (to a depth of several microns); in particular, lithium [McKubre 1993], **(e)** a highly dendritic layer of Pd with approximately 2-3 mm thick was visible at the electrode within the first 40-50 hours of electrolysis [Bockris 1993], **(f)** the foil (cells with Pd foil) is less productive than the powder (cells with Pd powder and Si powder) because of smaller surface area and the fact that the surface barrier is largely absent [Claytor 1993], **(g)** a peak attributable to HT appears and increases with the time in the final stages of ^{4}He production [Yamaguchi 1993], **(h)** the role of CaO layer in Pd complex [Iwamura 2006a].

The fundamental problems related to the premises of the model in relation to the compound CF materials will be developed in another paper presented in this Conference.

H. Kozima JCF21, **21-3**,

Neutron Energy Bands in the Compound and Composite CF Materials – Speculation on the Bases of the TNCF Model –

Hideo Kozima

Abstract

Using a phenomenological approach by the TNCF (trapped neutron catalyzed fusion) and the ND (neutron drops) models, we have given a unified explanation of the complex features of the cold fusion phenomenon (CFP). In the phenomenological approach, the necessary and sufficient conditions for the cold fusion phenomenon (CFP) has been established as the formation of the neutron energy bands (neutron bands) in the super-lattice of host elements and the hydrogen isotopes.

In this paper, the bases of the TNCF model are investigated in the rather complicated structure of the compound CF materials (multilayered materials and materials on substrates with interfaces) and composite CF materials (alloys and ceramics) by analogy with the electron energy bands (electron bands) in alloy semiconductors and at p-n junctions.

The neutron bands in the compound CF materials are investigated with reference to the electron bands in PN junctions. On the other hand, the neutron bands in the composite materials are investigated with reference to the characteristics of the electron bands in alloys at around symmetrical points in the Brillouin zone. The analogy between the electron bands and the neutron bands legitimates the use of the concepts of the neutron bands for investigation of the CFP in composite CF materials.

It is shown that the effects of the boundaries of the compound CF materials on the CFP are essential to induce the nuclear reactions in the CFP between the neutrons in the bands and nuclei at disordered positions generated by the thermal motion, the statistical distribution at a finite temperature, and the specific situation at around interfaces.

Comparison of AHE data between H2 and He runs for CNZ7rrr Sample

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Evaluation of anomalous heat effect (AHE) by the interaction of 3rd re-calcined nano-composite CNZ7 (Cu_1Ni_7/zirconia) sample[1,2] and H2-gas at ca. 300 degree C RC (reaction chamber) condition was studied by the comparison of same sample with H2-gas foreground and He-gas calibration runs. The sample was the third calcined CNZ7 sample, called CNZ7rrr (334 g), exclusively expecting relevant increase of integrated excess heat. AHE at elevated temperature was clearly measured with 36-59 W of excess thermal power level through the phase from the rise-up by heater on to 8 hours elapsed stage. Large increase (over 100 degree C cf. Helium run) of local temperatures was observed at saturated AHE power phase by the H_2-gas run with 235W heater power input. We also observed the increases of temperature at several points along the central line of RC. The most impressive time-point was of rapid increase after exceeding 300 degree C in the most effective condition zone near the W2 heater.

We make summary as: Comparison between H2 run and He run is effective to evaluate accurately the AHE excess thermal power, for different samples and in selected RC conditions; namely, heater input powers, heat capacity of RC (including test sample) and the situation of heat transfer from RC to outside. Another analysis was done for the increased temperatures of four RC points of RTD sensors with Helium run, to compare with data by H2 runs. From the engineering point of view, the more effective RC would be designed one with flat temperature distribution along the center line of RC, as much flat as possible. The comparison evaluation method of hydrogen run with helium run is effective not only for measuring the excess heat but also observing dynamic behavior of this kind of metal hydrogen energy along the temperature change.

Any negative effect of He gas for the sample in the RC was not observed till now, but detail examination should be confirmed from the several points, for example reliability and durability of sample, and any effect to the amount or the situation of the absorption and desorption of hydrogen into the sample.

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This is abstract to paper for JCF21 Meeting, Kyoto University, Dev.11-12, 2020

Thermodynamic analysis of Pd – H electrode: H/Pd > β_{min} during repeated cathodic and anodic electrolysis in an acidic solution

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Cold fusion experiments at ambient temperatures were conducted by electrolysis of heavy water on a Pd electrode. For a precise understanding of surface and cross-sectional morphology of deuterated Pd, *in situ* measurements of the electrode potential, dilation, and resistance of Pd electrode are of interest under well-controlled H absorption [1]. This study is a continuation of the reported works. We adopted the electrolysis method to conduct hydrogen absorption in glycerin-phosphoric acid accompanying *in situ* measurements of those physicochemical properties.

For the first C mode the *in situ* measured potential (E), dilation ($\Delta l / l$) and resistance ratio (R/R_0) exhibit the characteristic phase change: $\alpha \rightarrow \beta$ as a function of x: H/Pd ratio. Furthermore, the repeated C mode electrolysis was performed followed by potentiostatically controlled desorption discharge.

Hydrogen molecules can be absorbed in Pd metal or form a Pd hydride according to the reaction:

$$Me + x/2H_2 = MeHx + Q$$

where Me is Pd, a solid solution, or an intermetallic compound; x the molar ratio of H to Pd: H/Pd, and Q the heat of reaction.

In Pd-H system a solid solution referred to α phase is defined as solute H atoms occupy the octahedral sites randomly. The α phase regarded as primary solid solution changes to β phase due to, e.g. a difference between the radius of solute atom and that of solvent one. Like a substitutional solid solution, the solute atoms may be encountered by the disturbance, i.e. the disparity in size, the valence thereof and electronegativity. With an increase of H concentration, over the α single and $\alpha + \beta$ coexistence regions, the potential exhibits non Nerntian behavior: the potential deviates from a straight line whose slope is coincident with -62 mV/decade vs. the logarithmic scale of H/Pd. In these potentials, i.e. β single phase, the peak of the apparent molar volume was measured accompanied with the potential shift. Here, data of the potential and apparent molar volume changes might be analyzed using thermodynamic calculation as follows:

At first $\Delta_f G$ vs. T and $\Delta_f G$ vs. x diagrams will be estimated, hence it is necessary to collect and evaluate the thermodynamic parameters of Pd-H system obtained by metal-Hydrogen gas equilibrium method.

In the forgoing calculation, order-disorder transformation could be expected from the primary solid solution to some ordered phase or a precipitation of an intermetallic compound, similarly in the case of quenched alloys. Besides any substances estimated, it should be mentioned that the postulated structure is energetically more favorable than the starting one.

In this study H/Pd > βmin , the starting structure is known β single phase (H/Pd = 0.6) where the octahedral sites (½, 0, 0 ; 0, ½, 0 ; 0, 0, ½) of the fcc lattice are occupied by the hydrogen. It further postulates the transition for another interstitial site along diagonal at the ¼, ¼, ¼ or tetrahedral site. It is also essential to use precise lattice parameters and principal thermal constants whose experimental conditions, i.e. temperature, pressure, pretreatment of materials et al., might be suitable for the calculation.

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Simulation Study of Heat Conduction in Deuterium Desorption Experiment

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We have conducted deuterium absorption and desorption experiments using the Pd foil coated with metal membrane such as Pd-Ni, Pd-Zr, and Pd-Ni-Zr with fine-structure at the interface. In the experiments, we observed some peculiar temperature behaviors in desorption process. Short-period temperature fluctuation was often observed, lasting for 2-4 h at the beginning of the desorption phase [1]. Additionally, an instantaneous large heat evolution was occasionally found for Pd-Zr sample [2]. We estimated the heat energy balance in the unique phenomena considering the possible processes to specify the origin, especially for occurrence of a nuclear rection, and we have not confirmed significant heat of unknown origin so far [3]. We now still need to improve the calculation of the heat balance from the temperature variation of the sample to identify an anomalous phenomenon accurately.

In this study, we simulated how heat is transmitted in the sample and the circumstances as well as radiation heat when heat evolution occurs at a spot on the sample using the finite element analysis tool “ANSYS Mechanical APDL”, and the heat conduction and the radiant heat were evaluated quantitatively.

Our experimental setup, a sample placed in a stainless chamber, was modeled in the simulation [3]. Then, we investigated the transient behavior of the heat conduction with various conditions as well as the calorific value of the sample. In addition, the total amount of radiation heat was obtained from the simulation. This accurate evaluation of the heat transfer helps us specify a nuclear phenomenon in our experimental results.

References

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Laser Irradiation to D-Loaded Pd II

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A gas-phase experimental research in quest of condensed-matter fusion is underway by using multilayered deuterium-containing Pd plates. In our experiment, we in particular directly apply a bias voltage across the Pd sample to provide a current injection through Pd, to stimulate the nuclear reaction by Joule heating, also anticipating strong electrodiffusion or electromigration, in addition to the conventional deuterium diffusion induced by pressure/mass-concentration and thermal gradients.

The intensity and density of the triggering energy supplied to activate the nuclear fusion reaction are key factors to produce a smooth and reproducible initiation of the reaction. We previously proposed and numerically analyzed a scheme to provide high-density optical or electromagnetic energy to fusion-fuel materials by lasers and plasmonic field-enhancement effects, to significantly increase the reaction probability [1–3].

Our experimental setup is a stainless-steel-made, gas-phase, clustered reactor system including a deuterium-loading chamber, an electron-beam deposition chamber, and a reaction-analysis chamber. A high-resolution small-amu quadrupole mass spectrometer, two gas proportional neutron detectors, a Geiger-Mueller detector for α, β, and γ rays, and two liquid scintillators are equipped to the facility. In the present work, we have installed multiple kinds of lasers in the gas-phase D-Pd reaction system to irradiate the Pd samples, as an energetic stimulation support, potentially with a boosting plasmonic local field-enhancement effect. The lasers are 405-nm and 594-nm continuous-wave semiconductor lasers with powers of 50 and 30 mW, respectively, and a 1064-nm pulsed YAG laser with a peak power of 10 MW.

Previously we reported simultaneous observation of a sudden temperature increase with an overshoot and a neutron signal, accompanied with a clear signal of substantial-amount ^{4}He generation from the Pd samples as a shoulder peak on the D_2 peak, and a gas species with a mass number of three, via in-situ mass spectroscopy [4,5]. We present further analyses of our reaction system at the meeting.

This work was financially supported, in part, by the Japan Society for the Promotion of Science (JSPS) and the Research Foundation for Opto-Science and Technology.

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Optical observation on Anomalous Heat Generation from Nano-sized Metal composite with Hydrogen Gas

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Since 2015, Tohoku University has established a division of Condensed Matter Nuclear Reaction, and has been conducting research on anomalous heat generation phenomena using hydrogen and nano-sized metal composite. As a result, we have succeeded in observing the anomalous heat generation phenomena that cannot be explained by the chemical reaction[1]-[4]. On the other hand, the mechanism of the reaction has not yet been elucidated.

At JCF20, we discussed the following: When the novel nuclear reaction occurs, a region with very high energy density would be formed locally in the condensed matter. Then, emissions of low-energy photons can be expected as fast energy dissipation from the local high-energy-density spot. The wavelength range of low-energy photons is very wide; from X-ray to infrared radiation. In particular, we are interested in the spectrum of the visible light, because it may indicate thermal radiation from a local high-temperature region (hot spot)[5]. In the present work, we report on the measurement of visible light spectrum when anomalous heat generation occurs.

Our experimental process is as follows. First, we set samples in which Ni/Cu nano multilayer films are formed on Ni substrates by sputtering. These are fixed on both sides of a ceramic heater installed in the vacuum chamber. After baking, the sample is heated up and temperatures are measured for several heater inputs with a thermocouple placed inside the heater. At the same time, optical spectra are measured using a spectroscope (C10027-01; Hamamatsu Photonics K.K.). These are treated as reference data without excess heat generation.

Next, we introduce hydrogen gas into the chamber (about 200 Pa) and keep the temperature at 250℃ for 15 hours. This causes the sample to absorb hydrogen. Then, we evacuate the chamber and at the same time increase the heater input to raise the sample temperature. As a result, the absorbed hydrogen is discharged from the sample through the thin Ni/Cu layer. From this point on, measurements of the heater temperature and optical spectrum begin and continue, continuously for the temperature and every 5 minutes for the spectra. We evaluate excess heat generation by comparing these measurement results with the reference.

As a preliminary result, for the Ni substrate sample, there was no change in temperature and optical spectrum between the measurements with hydrogen absorption and without hydrogen (reference data). On the other hand, for the Ni/Cu nano multilayer sample, both optical emission rate and heater temperature of the hydrogen absorbed sample were clearly higher than those of the non-absorbed sample.

Details of the experiments and results will be reported.

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A theoretical study on the possible change of the phonon dispersion relation due to the nuclear reaction in two-dimensional lattice Ⅲ

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Abstract

In JCF19 [1], we showed the possible change of phonon dispersion relations due to the nuclear reactions at the uniformly distributed sites of crystalline solids. In JCF20 [2], we also showed the method how to detect the local change of lattice defects due to the nuclear reactions by introducing the theory of local modes described in the text book of C. Kittel [3].

In this study, we show the method how to detect the local change of lattice defect by generalized theory of local modes. This theory may provide evidences of the nuclear reactions in solids hidden in the old data of phonon dispersion relations.

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