

**6th International Workshop on Anomalies in
Hydrogen / Deuterium loaded Metals
*Pontignano (Siena) May 13-16, 2005***

**Prospects for Radio-active Waste Remediation
*A critical note on IJ Project***

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History

In July 2002 an article entitled *Elemental Analysis of Pd Complexes: Effects of D₂ Permeation* was issued on the Japanese Journal of Applied Physics (JJAP). Y. Iwamura, M. Sakano, T. Itoh of the Advanced Technology Research Center, Mitsubishi Heavy Industries Ltd. were the authors.

In 2002 again, an article with the same content, but entitled *Observations of Low Energy Nuclear Reactions induced by D₂ Gas Permeation through Pd Complexes*, was produced at the Ninth International Conference on Cold Fusion, Beijing, Tsinghua University.

The articles aroused a worldwide interest among Cold Fusionists. On December 2004 Iwamura was awarded a TEET grant.

At the *Tenth International Conference on Cold Fusion*. 2003. Cambridge, MA, T. Higashiyama, A. Takahashi et al. produced a paper entitled *Replication of MHI Transmutation Experiment by D₂ Gas Permeation Through Pd Complex*.

On 22nd January 2005 the economic Italian newspaper Il Sole 24 Ore issued an article by Dr L. Manusardi Carlesi: *Dal Giappone un passo avanti per rendere innocui i residui* (From Japan a step forward to make wastes harmless). The article gave an account on an Italian-Japanese Project aimed to the remediation of radioactive wastes from nuclear power plants, based upon Iwamura's claimed discoveries. Some important public and private Italian corporations and authorities would be involved in the project. The Italian public contribution would be of 16 millions euro in five years.

There is no information about the planning of a feasibility study about the project, probably because the involved corporations belong to the "believers" in cold transmutations through charged particles and can rely on political credit strong enough to bypass procedures, which are customary, especially when public funds are requested.

The IJ Project drew the attention of an informal discussion group in which CF and related topics had been treated for some years. It resulted soon that the matter was discussed almost only by physicists and that the contribution by chemists was badly lacking.

Meanwhile the 6th International Workshop on Anomalies in Hydrogen/Deuterium loaded Metals (Pontignano, Siena, 13 – 16th May, 2005) was promoted. The co-chairman of the Workshop, Dr William Collis, wrote these words to me:

I have not been following the recent email debate regarding the proposed Italian-Japanese project but it certainly seems to have provoked considerable interest in the international community. I believe you have played an important role in making our colleagues re-evaluate their positions. Science makes progress by making and responding to constructive criticism. I have therefore decided to dedicate the evening of Friday 13th to round table discussion "Prospects for Radio-active Waste Remediation". I think it would be appropriate if you were to mould your paper around this topic.

An open-minded attitude, sensible words, a kind invitation which deserved a positive answer.

The answer consists in a critical analysis of Iwamura and Higashiyama's papers, checking their chemical consistency.

The following note is addressed to people who know Iwamura and Higashiyama's papers; no details about Iwamura's transmutation device will be given.

Critical Note on the IJ Project

Experimental

Dr Iwamura and co-workers tried to load Cs, Sr and other elements on his Pd complex in three different ways:

Electrodeposition, Atom sputtering, Ion implantation

Let us have a look at the results of their trials

Electrodeposition

The description of the electrodeposition process is critical for the experiment and for the whole IJ Project

After forming a Pd complex, Cs or Sr was deposited on the surface of the thin Pd layer. Cs or Sr atom was deposited by applying an electric field to 1 mM CsNO_3 (with purity up to 99,9 %) or SrOD (with purity up to 99%) solution. A Pt wire (with purity up to 99,9%) was utilized as a counter-electrode. A 1 V negative voltage was applied to the Pd complex for 10 sec.

Referring to standard electrochemical data, we get:

Caesium Standard Oxidation Potential : 2,923 V

Strontium Standard Oxidation Potential: 2,890 V

It results that no electrodeposition is possible from an aqueous environment, less than ever by imposing a mere 1V across the electrodes. Chemical thermodynamics forbids that.

Palladium on the complex can only be spotted by tiny traces of Caesium and Strontium left behind from the very diluted solution.

At least in the original paper, where only the electrodeposition is proposed, no interaction between Caesium, Strontium and Deuterium could be proved, as there were no Caesium and Strontium to deal with but for traces, which do not justify the important presence of Praseodimium and Molibdenum claimed by the authors.

In a personal message, Prof. Takahashi confirmed the validity of Iwamura's electrochemical method:

A member (chemist) of Dr. Iwamura came to my laboratory to show us how to deposit Cs on surface of CaO/Pd layer of sample plate, based on "electro-chemical technique".

An important generalisation of the above remarks can be drawn:

Chemical thermodynamics forbids any electrochemical separation of Caesium and Strontium from radioactive wastes.

Iwamura's breaking of chemical thermodynamics can't be put aside, as it was displayed and confirmed many times in formal, public papers; based on this flaw, an experience was described, which should therefore be rejected as a whole. This conceptual fault, confirmed by Takahashi, discloses inexcusable uncertainties in mastering the fundamentals of chemistry.

After examining the inadequacy of the electrochemical way, a question arises:

Is a chemical separation of elemental or ionic Caesium and Strontium from aqueous nuclear wastes possible from a technical point of view?

In the last few years it seems that CSSX (caustic-side solvent extraction) and sequestering organic molecules like *calixarenes* achieved the goal. But luckily in this chemical option there is no need of transmutations. The small volume of the nuclear poison can be made harmless by conventional ways, vetrification and the like.

Atom sputtering, ion implantation

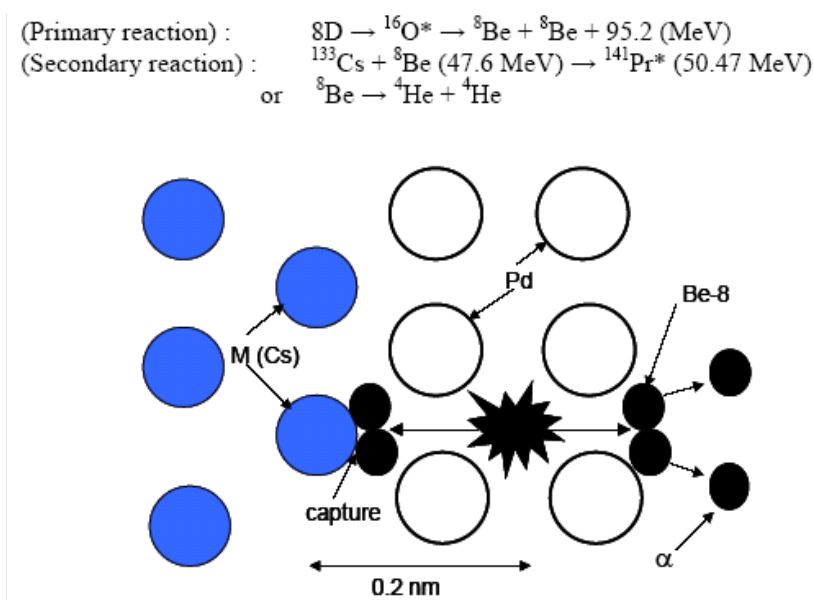
Recent claims on the greater fitness of these techniques change nothing: while an electrochemical technique could be applied on a large scale, atom sputtering or ion implantation are not suitable to be scaled up. They may only be auxiliary, laboratory tools. Nuclear wastes typically require simple and reliable standard conditioning techniques, owing to their bad handiness.

Theoretical views

Dr Iwamura does not provide his experiment with theoretical considerations; the task is left to Prof. Takahashi who duplicated Iwamura's experiment together with Higashiyama's team. Takahashi's theory, called “8 D nuclear fusion”, is unique in explaining Iwamura's transmutations, as far as I know.

In short:

Two high energy ^8Be are produced by octahedral resonance fusion of eight deuterons in the Pd lattice. Beryllium nuclei can either decay into 2 ^4He or collide with Cs producing Pr in only one step. The same happens with Sr as a target.



This theory is brand new; it has no links with classical Metal Science and Nuclear Chemistry.

Takahashi's reactions require deuterons, not deuterium atoms. All classical and recent views in Metal Science claim that hydrogen and deuterium are present in metal lattice as ions subjected to an electronic screening, just like the components of other metal alloys.

Let me introduce only one quotation.

Prof. Renzo Valentini, University of Pisa, expert in Hydrogen-metal alloys (2005):

L'idrogeno, ed i suoi isotopi, vengono sempre assorbiti in forma atomica nei metalli (non in forma ionizzata). Questo vale anche per il Pd. (Always hydrogen and its isotopes are absorbed in their atomic (not ionic) form in metals. This also applies to palladium).

An indirect but important clue against considering deuterium in palladium lattice in an ionic state is given “in negativo” by Fleischmann, Pons, Preparata (*Possible Theories of Cold Fusion, Il Nuovo Cimento, gennaio 1994*) who maintain:

...bosonic character of D^+ as opposed to the fermionic character of H^+ and T^+ , the Pauli principle restricting the configuration space of H^+ and T^+ but not of D^+ .

Were this statement true, so would the state diagram Pd/D be completely different from Pd/H

diagram, but this doesn't happen (M. H. Maxelon, *Segregation von Wasserstoff an Versetzungen in Palladium* (Hydrogen segregation inside displacements in Palladium), Göttingen 2000).

Atomic deuterium, not *Bosonic* D^+ , seems to be present in Palladium lattice.

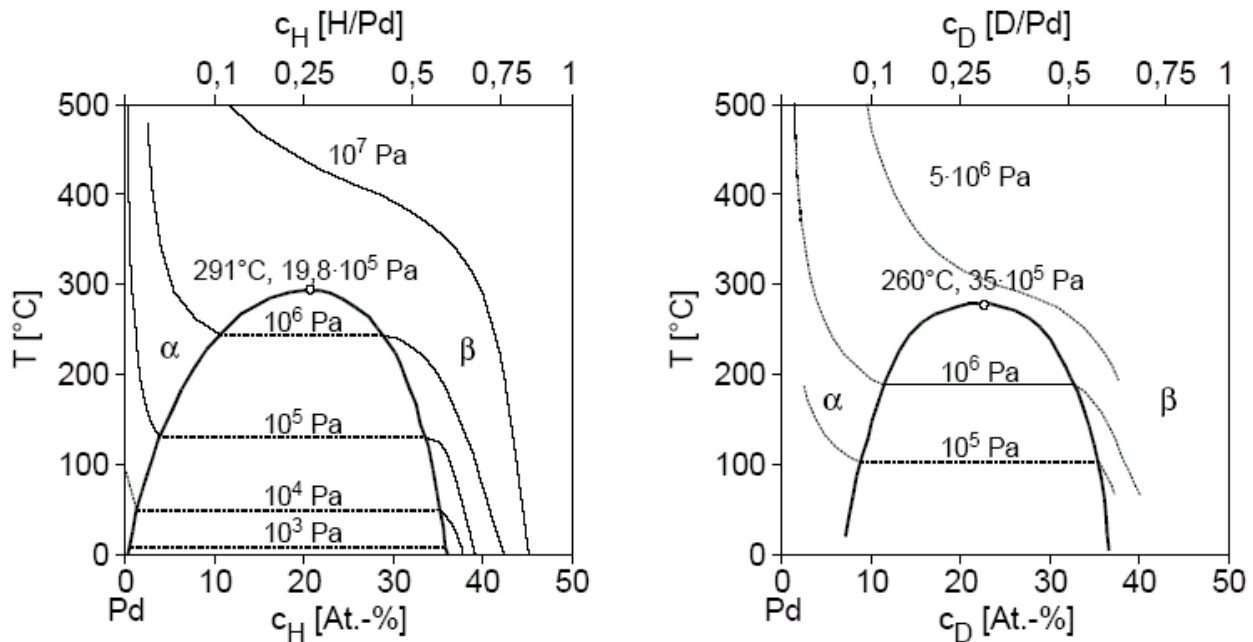


Abbildung 2.1: Phasendiagramme der Systeme Palladium-Wasserstoff und Palladium-Deuterium.

Fig. 2.1: Phase diagrams of the systems Palladium/Hydrogen and Palladium/Deuterium

The production of hot oxygen nuclei from deuterons has no connection with all notions developed along hundred years in nuclear and radiochemistry. Multibody interactions are brought in, conflicting with chemical kinetics, to which even a three-body collision is practically impossible. While the reality of the $dd \rightarrow {}^4\text{He}$ reaction in palladium lattice is still under examination, Takahashi introduces a new reaction, much more questionable because of its multiplicity.

Resonant fusion is normally obtained in modern crossed beam accelerators (LEP, CERN). In accelerators only two crossing beams are tunable; so far, more than two, less than ever eight, is beyond any technical reach.

Has perhaps the expression “resonant fusion” a different meaning in Takahashi's model?

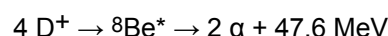
Nuclear reactions among charged particles are considered typical threshold reactions, as particles must belong kinetic energy to overcome the coulombian barrier. For example the reaction dd needs an activation energy around 200 keV.

Even considering tunnel effects, we must cope with the coulombian barrier, especially with multiproton nuclei.

Where does the collision energy come from in Takahashi's model?

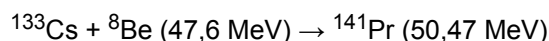
Prof. Preparata was sarcastic towards multy-body resonant fusion (Fusion Technology, Vol. 20, 1991):

No surprise that we can but admire in awe equations such as:



How high a Coulomb barrier does one have to climb before one can reach the mythical ${}^8\text{Be}^*$ nucleus? I believe that exposing these kind of fallacies that are being pursued in the field of cold fusion is absolutely necessary if we wish to prevent it from becoming esoteric, thus drifting away from scientific realm.

The major objection is reserved to the following reaction, Takahashi defines “capture”:



How can Takahashi pretend that a nucleus with 55 positive charges “captures” a nucleus with 4 positive charges? We can allow for the kinetic energy derived from the fission of the hot oxygen, but everybody can argue energetics is not respected.

Nuclear reactions usually occur step by step; transuranic build up in nuclear reactors is a typical example. In Takahashi's model a multibody projectile is built in advance. This seems to be done in order to bypass the problem of the half-lives and cross sections of the intermediate nuclei, which could hinder or stop the reaction chain towards a stable nucleus.

Definitely nuclear chemistry cannot accept Takahashi's model.

In my opinion, should Iwamura's transmutations hang on Takahashi's model, they would never occur.

Fast Neutron Activation Analysis (FNAA)

Prof. Takahashi and his team tried to detect ^{141}Pr by FNAA, using the 14 MeV neutrons produced by the Japanese FNS (Fusion Neutronics Source)/JAERI accelerator. The analytical method deserves some quantitative remarks:

- Total estimated quantity of ^{141}Pr produced by transmutation:
 10^{14} atoms, say 20 ng.
- Detection limit for ^{141}Pr , using NAA: 30 ng, for a thermal neutron flux of $10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ (SPNR).
- Fast neutron absorption cross section: $2,5 \cdot 10^{-2}$ barn
- Thermal neutron absorption cross section: 10 barn
 For the same neutron flux, NAA is 400 times more sensitive than FNAA.
- Total neutron output of FNS: 10^{12} n s^{-1} .

Neutrons crossing the 625 mm^2 complex surface can be only a fraction of the total output.

Conclusion:

detection limits for ^{141}Pr are at least 3 orders of magnitude above the estimated quantity of Pr produced.

FNAA is definitely unfit for detecting small quantities of Pr.

NAA would work better, even if 10 barn remains a modest quantity to get fine results.

FNAA is suitable for the quantitative determination of light elements, such as Ca and O, which are just present in the complex and contribute heavily to the background noise.

Conclusions

Nuclear and radiochemistry have always been strongly committed in radioactive waste remediation. Their proposals are based on the huge store of knowledge collected over the years. Some suggestions about “academic” transmutations of nuclear wastes, taken from a recent nuclear and radiochemistry textbook, are reported in the following page.

No surprises in reading it; natural laws and common understanding are respected.

21.11.5. Transmutation

The fission products ^{90}Sr and ^{137}Cs can be transformed into shorter lived or stable products by charged particle or neutron irradiation. Charged particle irradiation would be very expensive, and irradiation by reactor neutrons would produce almost as much fission products as are destroyed. Therefore the use of intense accelerator driven spallation neutron sources for transmutation by n-irradiation has been suggested. If controlled thermonuclear reactors (CTR) are developed, their excess neutrons could be used for ^{90}Sr transformation, but less efficient for ^{137}Cs .

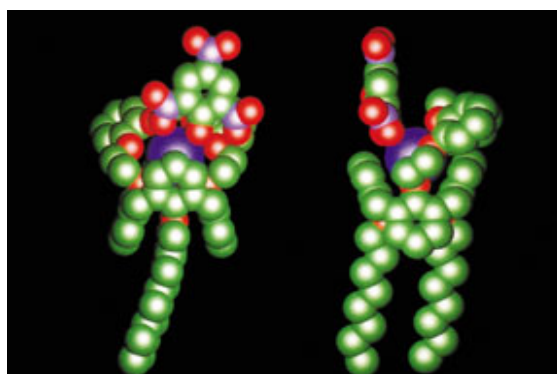
In the long term (≥ 600 y) the actinides dominate the risk picture. Continuous neutron irradiation of the actinides finally destroys all of them by fission (cf. Fig. 16.3). The annual production of americium and curium is ~ 5 kg in a 1000 MW_e LWR, but considerably less in a FBR. Thus if pins of these elements are inserted in a FBR, more americium and curium is destroyed than formed; it is estimated that 90% will have been transformed into fission products after 5-10 y. In the future, CTRs could be used for the same purpose. As an alternative it has been suggested to leave the americium and curium in the uranium returned in the LWR cycle. Wastes from transmutation processes will contain some amount of longlived nuclides, thus a safe final repository is still needed.

G. Choppin, J-O. Liljenzin, J. Rydberg, *Radiochemistry and Nuclear Chemistry* Butterworth, Heinemann, 2002 (revised December 2004)

The very recent prospects of radiochemical approach to the remediation of nuclear wastes, leading to very small volumes of nuclear poison, make useless any further treatment: vetrification of sequestrated nuclides is an excellent, cheap containment tool.

Modern theoretical chemistry, based on Feynman's QED and on minicomputers, can build specially conceived organic molecules, stable towards nuclear radiations, able to trap only one ionic species and release it, when chemical environment is made to change, so retrieving the expensive reactant. Selectivity, small volumes of nuclear poison, reliability, cheapness are the valuable characteristics of these remediation techniques.

In the following figure a Caesium ion (dark violet) is trapped in a calixarene-crown molecule.



These molecules have been synthesized at the University of Parma (Italy) by Prof. Ungaro and his team. Lots of patents are protecting molecules and processes. The properties of calixarene-crown molecules are studied in Cadarache (CEA, France) for use in extracting Caesium.

Iwamura's physical method, involving electrochemistry, ion implantation and atom sputtering and eventually transmutations seems to be outside recent trends: too complicated to become an

industrial process; not defined in its chemical sides yet; lacking any theoretical support .

Moreover the production of the basic reactant, deuterium, is very energy demanding. Assuming that Takahashi's model is correct, how many berillium nuclei decay into alpha particles, so wasting deuterium? Building a nuclear plant; decommissioning it at the end of its working life; remediating the nuclear waste using deuterium can lead to a global negative energy balance (or not positive enough).

In Italy radiochemistry needs financial support and fresh image; many important radiochemical plants were dismantled after the referendum against nuclear power in 1986 (Rotondella, Medicina, among others); only few students are attending radiochemistry courses, as radiochemistry laboratories have almost disappeared.

Like reprocessing of nuclear fuels, nuclear waste remediation is mainly a job for radiochemists and chemical engineers. Physicists offer a valuable support of course.

This is a little polemical, but friendly note, because I believe that in this section dealing with the Prospects of Radio-active Waste Remediation, physicists are more numerous than chemists, not to say radiochemists.

Have physicists more political audience than chemists? I hope not.