Synthesis Of A Copper Like Compound From Nickel And Hydrogen And Of A Chromium Like Compound From Calcium And Deuterium

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ABSTRACT

A working hypothesis is presented that aims to explain results observed in the LENR-CF field. This hypothesis is based on a novel conjecture: a very sizeable increase of the strength of gravitation at pico-meter distances. Experiments designed to confirm (or deny) this working hypothesis are described.

Introduction

Various phenomena have been observed when hydrogen isotopes are brought in contact with metals or submitted to the action of an electrical discharge. Among them, most striking are the following:

- a positive difference between the energy released by a system under experiment and the energy it receives from the outside has been observed for instance in the electrolysis of heavy water with palladium electrodes (Fleischmann and Pons). This has been called "excess energy."
- elements with an abnormal isotopic composition have been observed to appear in some systems (electrolysis, deuterium permeation through palladium, glow discharge…) during and sometimes after the experiment. (Iwamura, Miley, Savvatimova…).
- according to various teams (Oriani, Storms and Scanlan) nuclear radiations are emitted in some systems (electrolysis, glow discharge…) during and sometimes after the experiment.

Given the very low level of "reaction products" appearing in the experiment, when compared to the level of "excess energy" observed, these phenomenon have been ascribed to a special kind of nuclear reactions (Cold Fusion: CF, Low Energy Nuclear Reactions: LENR). These reactions would emit several orders of magnitude less radiation than expected from the "excess energy" or the amount of "reaction products" observed.

A series of experiments have been run to shed some light on the field. They have been designed to confirm or deny a conjecture that was previously presented [1] and that might explain the whole features of the CF, LENR field. They imply contacting hydrogen

isotopes with various metals, under well defined experimental situations. That could result in the synthesis of unusual chemical elements, formed by a novel type of binding between the metal nucleus and the hydrogen isotope (a bond strength ranging from hundreds of electron volts to several thousand electron volts). The results will be presented and discussed in relation to this conjecture.

Experimental strategy

From an operational point of view, these reactions (assuming they exist) are very difficult to trigger. A macroscopic effect (sizeable excess energy) is seldom achieved. This might be the consequence of a relatively high level of energy required for those unknown reactions to occur (5 to 10 eV). Such levels of activation energies are indeed found in metal lattices or in gaseous plasmas. These reactions usually result in the appearance of reaction products in trace amounts and/or the emission of nuclear radiations, without measurable "excess energy." It has nevertheless been thought that the microscopic features of these products could well provide guidance for better controlling the macroscopic effect (excess energy). As will be seen, the basis of the experimental approach is to characterize the trace elements produced. It is thus of paramount importance that these traces be perfectly controlled in the starting material. Various situations have thus been studied to choose the one that satisfies best this condition. In this series of experiments, 3 types of situations, in which hydrogen isotopes are in contact with metals have been studied or are under study:

- Situations where hydrogen is in contact with nickel. Various experiments have shown that in these conditions excess energy and/or "transmutation" of nickel into copper occur [2],[3]. Hydrogen in contact with nickel during the reduction of nickel chloride (**system I**) and the hydrogen in contact with Raney nickel (**system II**) have been studied. It will be seen that only hydrogen reacts in that case.
- Situations where hydrogen is in contact with calcium (in the form of calcium carbonate or calcium oxide). Palladium, present in the experiment yields atomic hydrogen (deuterium). This configuration is intended to reproduce experiments presented in [4], where strontium is transmuted into molybdenum. The approach was to contact hydrogen with a LINDLAR type of catalyst (**system III**). It will be seen that only deuterium reacts in that case.
- Situations where hydrogen, activated in a low pressure ozonizer is in contact with metals, are under study.

The following 3 systems have been studied:

- I   Reducing nickel chloride by hydrogen isotopes
- II   Hydrogen isotopes in contact with Raney nickel

- III Hydrogen isotopes in contact with Lindlar catalyst (Palladium on calcium Carbonate)

Experimental description

NICKEL

System I
A number of studies (particularly with regard to batteries) have been carried out with the objective of finding a process yielding finely divided nickel.

Such a process is described in [5]. Basically, nickel in nickel chloride is reduced to its metallic state by hydrogen, according to:

$$NiCl_2 + H_2 \rightarrow Ni + 2HCl$$
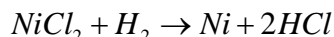
As such, the reaction proceeds at 450°C, yielding particles with mean diameter 2.5 µm (see [5]). The addition of 0.1% weight palladium allows a lowering of the reduction temperature to 380°C, together with a reduction to 0.4 µm of the mean diameter (see [5]).

Experiments have been carried out using $NiCl_2, 6H_2O$ (Rectapur from PROLABO), containing less than 5 ppm copper, to which 0.1% weight palladium was added according to [5]. The reduction was carried out in a vertical Pyrex reactor, heated to 380°C, the hydrogen isotope flowing through the chloride to be reduced (see Reactor I, figure 1).

System II
Raney nickel is a well known catalyst used for hydrogenation. It is prepared by dissolving (by sodium hydroxide), the aluminum contained in an aluminum/nickel alloy. This yields finely dispersed nickel particles, supported on an alumina matrix. PROLABO Raney nickel 25 820.230 was used. The catalyst was placed in an alumina boat, inside a metallic reactor (see reactor II, figure 2). It was first dried and activated by heating to 450°C under vacuum. It was then placed in contact with a hydrogen isotope (hydrogen or deuterium) at 450°C, under a pressure of 5 bars, for 2 hours.
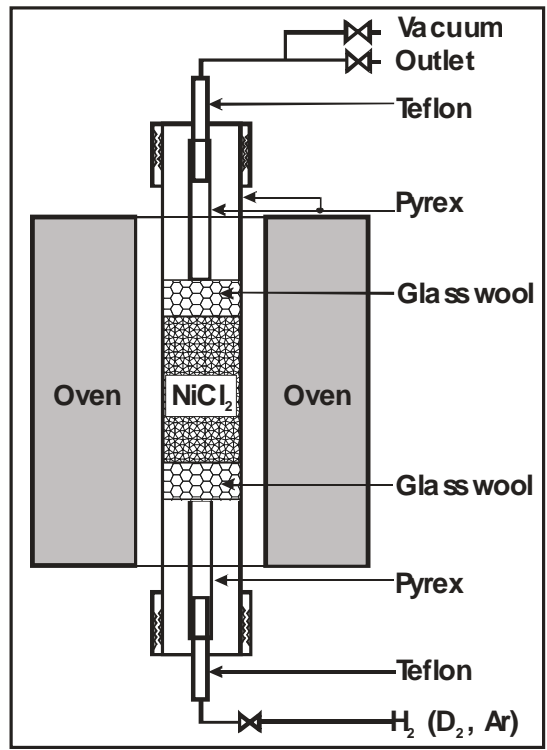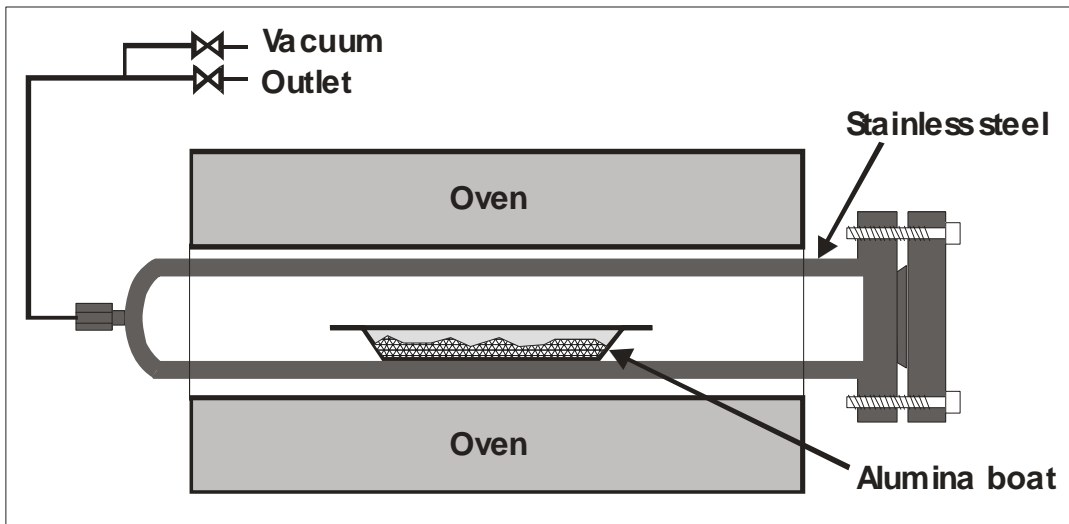
Fig. 1. Reactor I

Fig. 2. Reactor II

CALCIUM

System III:

Lindlar catalysts were developed to selectively hydrogenate acetylene bonds. Palladium is deposited on alkaline-earth carbonates as nano-particles. Palladium loading between 5 and 10% weight are used. Both a commercial catalyst (Alfa-Aesar palladium 5% weight on calcium carbonate) and a homemade one were used. Experiments were run after de-carbonation of the catalyst, thus yielding the CaO-Pd system. Hydrogen isotopes (hydrogen and deuterium) were placed in contact with the de-carbonated catalyst under various temperature and pressure conditions, in Reactor II.

Characterization of the processed samples

As mentioned in the introduction, the synthesis of novel chemical elements is suspected in the CF LENR field. To get insight on the features of these new elements, their outermost electronic layer has been explored by ICP-AES (Varian) available at the laboratory in CNAM and their nucleus by ICP-MS available outside.

All samples have been dissolved in hot aqua-regia. Nickel from the reduction of nickel chloride and Lindlar catalyst were completely soluble, whereas a residue (alumina) was left with the Raney nickel.

Main results

NICKEL

Results are summarized in Table 1 below. It can be seen that the copper content (in mgCu/gNi) is always higher for treated samples when compared to blank ones (RATIO mgCu/gNi), when hydrogen is used. One sample submitted to the action of deuterium shows no variation of the RATIO mgCu/gNi being within the error bars.

Table 1. Ratio of mgCu/gNi processed samples compared to blanks

		Reactor I					RATIO
		AES			MS		AES/MS
		Gas			Gas		
Nickel Type	Exp. No.	Hydrogen	Hydrogen	Deuterium	Hydrogen	Deuterium	Hydrogen
RANEY	1	1.976					
RANEY	2	2.102					
NiCl$_2$ red.	3		2.833				
NiCl$_2$ red.	4		**3.089**		**1.777**		**1.738**
NiCl$_2$ red.	5			**1.186**		**0.944**	**1.256**
NiCl$_2$ red.	6		2.636				
NiCl$_2$ red.	7		1.420				
NiCl$_2$ red.	8		1.842				
Mean value		2.039	2.364				
Standard Deviation		0.089	0.704				

Samples from experiments 4 and 5 (and a corresponding blank), were then analyzed by ICP-MS. It can be seen that ICP-MS sees significantly less copper than ICP-AES for experiment 4 (hydrogen), the RATIO AES/MS being 1.738. In contrast, experiment 5 (deuterium) is within the errors bars: no excess copper appears in the treated sample; ICP-AES and ICP-MS sees the same amount of copper (RATIO AES/MS being 1.256). This conclusion is in line with what was expected (see below for the case of calcium and the discussion of the conjecture)

ICP-MS did not reveal any significant variation of the isotopic composition of the copper found in the hydrogen treated sample compared to that of the blank one. An important increase of the $_{29}^{63}Cu$ isotope was indeed expected (see below the case of calcium for more detailed explanations). It cannot be excluded that the model presented in [1] is too simple to correctly predict the rate of reaction of the various isotopes involved (indeed reference [3] indicates an increase of $_{29}^{65}Cu$ compared to $_{29}^{63}Cu$). More trivial reasons could also play a role to explain this result:

- Conditions of the ICP-AES and ICP-MS measurements. Matrix and nebulization effects could play a role. This could be better understood with more elaborate (and more costly) analysis: standard addition in the case of ICP-AES, isotopic dilution for ICP-MS.
- Heterogeneity of the samples of starting material. Indeed, the nickel chloride to be treated is obtained by evaporation, and one cannot exclude a selective crystallization

of the trace amounts of copper. The case of Raney nickel is even worse, because an insoluble residue of alumina remains after the attack by aqua-regia.

In view of these problems, it was decided to switch to a more favorable system, prompted by the results of [4].

CALCIUM

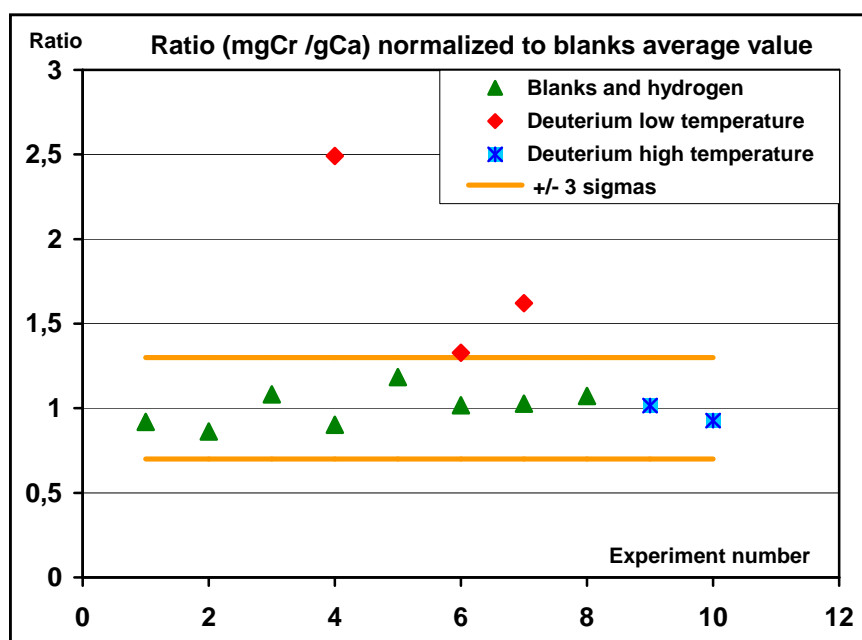Results are summarized in Fig. 3 below.

Fig. 3. Ratio (mgCr/gCa) normalized to blanks, average value

The calcium and chromium contents of the samples (respectively in g/l and mg/l) were measured by ICP-AES and the RATIO mgCr/gCa calculated. 10 experiments gave a comparable RATIO, yielding an average value with which all results were normalized. The lines "average value of the blanks ± 3 sigma" are shown in Fig. 3.

Various operating conditions were used:

- De-carbonation temperature.
- Hydrogen (deuterium) contact time.
- Hydrogen (deuterium) pressure.
- Blanks were run with argon, in comparable conditions and the RATIO mgCr/gCa was also determined on the untreated catalyst.

It can be seen from Fig. 3 that three experiments are beyond the 3 sigma limits (note that 2 experiments can have the same number on the graph. They are indeed different; it is just a matter of presentation of the graph).

Experiments with hydrogen, argon, deuterium, high temperature (450°C) and untreated catalysts are within the 3 sigma limits. In contrast, experiments with deuterium at low temperature (130 to 150°C) are beyond these limits.

No attempt as yet been done to measure chromium by ICP-MS. The results expected will be discussed in the following section. For the time being, the work on the system CaO-hydrogenating metal is directed in two directions:

Synthesize Lindlar type catalysts starting from calcium compounds having the lowest possible chromium content.

Submit Lindlar type catalysts (as a very thin layer spread on one of the Pyrex electrodes of an ozonizer), to the action of an ozonizer discharge in various gases (deuterium, hydrogen, argon…). This experimental set-up is very clean: the discharge taking place between 2 Pyrex dielectric barriers only sees Pyrex and the catalyst to be treated, thus avoiding pollution of the samples to be analyzed after the experiment.

Conditions of the discharge currently under study are:

 - Gas pressure of the gas 1 to 50 mb
 - Temperature 50 to 150°C
 - Distance between electrodes 5 mm
 - Voltage 1500 to 2500 V
 - Current 2 to 40 mA
 - Frequency 5 to 15 kHz

Under these conditions, a sizeable amount of H^+, D^+ or Ar^+ with energy of around 10 eV will impact the LINDLAR type catalyst.

Interpretation of the results

In order to make clear what is expected, according to the conjecture that will be discussed below, the case of calcium is summarized in Table 2.

Table 2. Isotopic composition of calcium

ICP – AES				ICP - MS			
ISOTOPE	COMPOSITION (%)	MASS	ADDITION OF 4 D	COMPOSITION (%)	MASS	ISOTOPE	COMPOSITION (%)
Ca 40	96.941	39.962591	48.018591		47.954033	Cr 48	
Ca 42	0.647	41.958618	50.014618	0.647	49.946046	Cr 50	4.345
Ca 43	0.135	42.958766	51.014766		50.944768	Cr 51	
Ca 44	2.086	43.95548	52.01148	2.086	51.940509	Cr 52	83.79
				0	52.940651	Cr 53	9.5
Ca 46	0.004	45.953689	54.009689	0.004	53.938882	Cr 54	2.365
Ca 48	0.187	47.952533	56.008533		55.940643	Cr 56	
100				2.737			100

In the first part of Table 2 (ICP-AES) the isotopic composition of calcium and the mass of the corresponding isotopes is given (columns composition and mass). In column "addition of 4D", the mass of hypothetical compounds, obtained by the addition of 4 deuterons to the various calcium isotopes is given. If the conjecture is verified (see below), ICP-AES, used for determining the calcium content of a sample, will see all these hypothetical compounds, because they all have the same outermost electronic layer.

In the second part of Table 2 (ICP-MS), the masses of known chromium isotopes (both stable and unstable) are given (in the Mass column). In order to determine the chromium content of a sample by ICP-MS, the amounts of isotopes 50, 52, 53 and 54 are measured. If the conjecture is verified (see below), the amount of chromium like compounds, coming from the various calcium isotopes and seen by ICP-MS will be much lower than the amount of these compounds seen by ICP-AES (2.737 compared to 100). Moreover, an upward shift of masses will be observed.

In other words, the ratio mgCr/gCa for a treated sample will be higher when measured by ICP-AES than when measured by ICP-MS.

More generally, when measuring the amount of converted products, there will be a discrepancy between the results obtained by ICP-AES and those obtained by ICP-MS. This was observed for the conversion of nickel to "copper" (see above).

The pico-chemistry conjecture

This conjecture has been presented in [1]. The following basic hypothesis is made: at distances within atomic dimensions (say 100 pm as an order of magnitude), there is a considerable increase of the strength of gravity, up to a level where the gravity attraction between the nucleus of an atom A and a proton (or deuteron) can counter balance the electrostatic repulsion between them. An (over-simplified) calculation points to the possibility of bound states between A and the proton (or deuteron), at distances well within the electronic layers of A. The enthalpies of reaction in this new chemistry (pico-chemistry) would in the range of hundreds of electron volts up to several thousand electron volts (partly emitted as X-Rays), whereas the activation energies would be tens of electron volts, which explains why it is difficult to trigger these reactions. In the case of deuterium, this initial pico-chemistry reaction could be followed by a true nuclear reaction, the deuteron reaching the nucleus of A and reacting with it, thus yielding a transmuted nucleus and various nuclear radiations (α, β, γ). The conjecture and its consequences are summarized in Fig. 4 below.
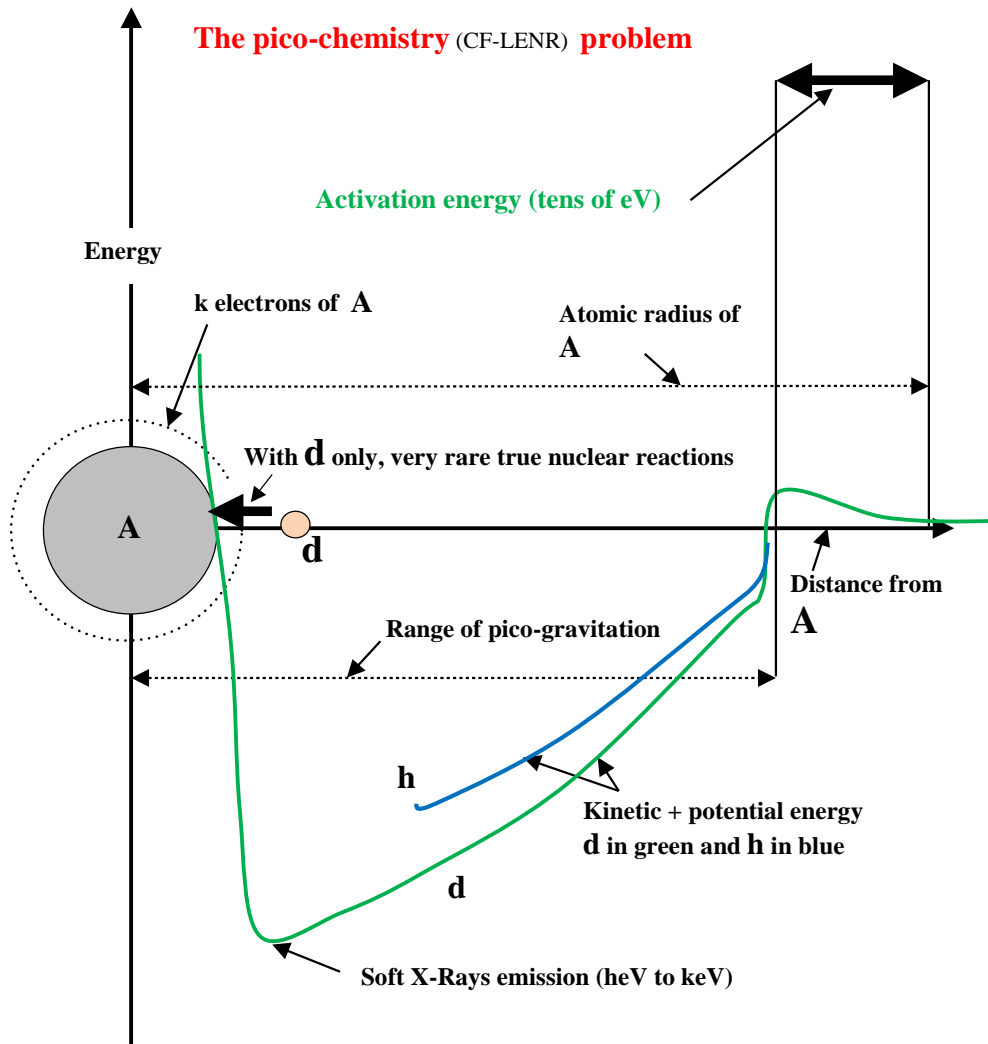
Fig. 4. The pico-chemistry (CF-LENR) problem.

As can be seen from Fig. 4, the pico-chemistry conjecture could explain:

The production of energy from deuterium and/or hydrogen (depending upon the treated atom A).
The emission of X-Rays.
The emission of nuclear particles (α, β, γ), in very small amounts.
The appearance of transmuted products.

Conclusion

The working hypothesis presented in this paper, leads to clean, simple and reproducible experiments that will confirm (or deny) the pico-gravity conjecture. If the results of these experiments are positive, a novel field of chemistry will open. It is proposed to call this new field "pico-chemistry". Pico-chemistry enthalpies of reaction (in the range of hundreds of electron volts to several thousand electron volts) could provide a new source of energy. A massive graviton, with mass between 25 to 50 keV, would be associated with pico-chemistry.

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