

Unknown Nuclear effects and Abnormal Isotopic Compositions

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I - Introduction

LENRs (Low Energy Nuclear Reactions, with a threshold lower than 1 keV/atome, even than 1 eV/atome, to compare with the "traditional" threshold of MeV/atome) are scientifically highlighted since about fifteen years in many experiments known as "Cold Fusion" experiments.

One of the arguments of the existence of these reactions (which we will call in a broader way "Unknown Nuclear Effects" or UNEs) in this type of experiments rests on the presence of elements whose Isotopic Composition (IC) strongly deviates from Standard IC (SIC). One qualifies then these ICs of Abnormal Isotopic Compositions (AICs).

First of all, we will specify the differences existing between SICs, Non Standard Isotopic Compositions (NSICs), well known physical phenomenon used in various fields of studies, and AICs.

Then we will see that if the researchers in LENRS are confronted, with great satisfaction, with AICs, they are not the only ones. Indeed, physicists of the atmosphere, cosmochimists, volcanologists, geologists, archaeologists, ..., all are confronted with problems of AICs.

But for these researchers, satisfaction yields the step to embarrassment.

II – Standard Isotopic Compositions, Nonstandard and Abnormal

We will limit our matter to the stable isotopes for these 3 categories of ICs.

The SICs are ICs which one finds on ground or in the solar system (sun, moon, planets...), inherited from the proto-solar nebula, itself even sown in isotopes created by stellar nucleosynthesis in the preceding generations of stars.

The NSICs are well known, indexed and explained by the following physical phenomena:

- isotopic fractionation,
 - decay of radioelements (explaining the NSICs of terrestrial Pb in particular),
 - nucleosynthesis by spallation induced by cosmic rays,
 - contribution of various stars with different isotopic signatures,
- and marginally, nucleosynthesis carried out by an Oklo type natural nuclear fission reactor.

The most common process is the process of isotopic fractionation: a physical process - evaporation, condensation, diffusion, thermal diffusion, gravity, phase transitions, exhaust of a planetary atmosphere, photolyses... - or a chemical process can support an isotope heavier or lighter, which leads to an IC which deviates from the SIC.

One quantifies the isotopic variation by an Isotopic Delta noted δE (δD , $\delta^{18}O$...) expressed in 1 part for 1 000 compared to the most abundant isotope. Isotopic variations δE maximum met in the NSICs are indeed weak, about 10 per thousand (i.e. 1 %), except for C, N and O for which they can reach 10 %, of S, Ca and Pb for which they can reach 20 % and of H, He and for which they can reach 100 % [1, 2].

Because isotopic fractionation is worth especially for the light isotopes (let us say $A < 40$ i.e. Calcium), a difference in mass of 1 for heavier isotopes quickly becoming negligible.

These NSICs are largely used by the scientific community like physical indicators of phenomena: variation of 3% of the presence of ^{18}O in polar carrots connected to the global temperature, variation of 3 % of the presence of ^{13}C in Chlorophyl connected to the diets of herbivores and carnivores, variation going up to 23 % of the presence of ^{204}Pb in the grounds to discriminate between endogenous lead and industrial lead...

The AICs, on the opposite to the NSICs, are characterized by the 2 following facts:

- the mechanisms called upon for the NSICs are not implied,
- the isotopic variations are very strong and reach 10 000 per thousand for certain elements, even more.

It is necessary to underline here that researchers having published compilations [1, 2] mention the existence of AIC measurements. For example, those obtained in geological samples for Ar, Kr, Rb, Zr, Mo, Ru, Pd, Ag, Cd, In, Sn, Sb, Te, Xe, Ba, Sm, Eu, Gd, Dy, Er, Yb, Lu and Os [1]. But these measurements are systematically rejected, supposed to be sullied with errors, even if those are not identified.

Hence, one understands that NSICs and AICs constitute 2 categories of very different results.

We now will see that the AICs are measured in many and various study fields.

III - AICs met in LENRs experiments

The first experiments of Cold Fusion carried out in 1989 reflect in obviousness, beyond the appearance of an excess heat whose intensity could not be explained by chemical reactions, the appearance of Helium (^4He) and, more marginally, of Tritium (the appearance of ^3He was measured only several years afterwards), coming from D-D fusion.

In 1991, Pr Matsumoto of the Hokkaido University was the first researcher to bring back the observation of new elements produced by LENRs process : Na, Mg, Al, Si, P, S, Cl, Ar, K, Ca, Ti, Cr, Fe, Ni, Cu, Zn, Ru and In [3].

The presence of rare gases (Ne and Ar) and of not very widespread elements (Ru and In) militated for the signature of LENRs. But for widespread elements such as Al, Si, Ca..., the assumption of a contamination could not be still excluded.

It is to dismiss this assumption that, in 1992, Dr. Karabut of the Russian laboratory LUTCH measured the radioisotopes of the new elements produced by the LENRs, namely Sr and Rh [4].

It was him also, which, since 1994, was the first to measure ICs of stable isotopes: Li, B, Si, K, Ti, Fe, Zr, Mo and Ag [5].

Since, in LENRs experiments, more than thirty elements presenting AICs were measured: Li, B, C, O, Na, Mg, Al, Si, S, K, Ca, Ti, Cr, Fe, Ni, Cu, Zn, Br, Kr, Zr, Mo, Ru, Pd, Cd, Xe, Hf, Ir, Pt, Hg, and Pb.

It would be tiresome to review all these results. Let us quote for example [6].

It is significant to underline here the universality of this type of results. Indeed, if in the first years, the researchers concentrated their efforts on the electrolytic cells with heavy water and palladium cathode, today, the results are obtained for a large variety of configurations of experiment:

- various states of the matter for hydrogen penetrating metal: liquid (hydrogenated electrolyte) but also gas, plasma and ion beams,
- various metals: Pd but also Ti, Co, Ni, Sr, Cs, W, Pt, Au, Pb, U,...
- deuterium but also protium.

One notes a constant in these experiments: the presence of Hydrogen (protium or deuterium) penetrating a metal, i.e. a condensed medium, in which these nuclear reactions occur. We will thus call these reactions Condensed Hydrogenated Medium Nuclear Reactions (CHMNRs).

To summarize the results of these measurements, let us say that:

- for certain elements, IC is close to or identical to the SIC,
- for other elements, IC is an AIC. Examples : factor 32 on the ^{33}S , factor 100 on the ^{57}Fe , factor 20 on the ^{65}Cu , factor 15 on the ^{202}Hg ...
- these AICs are generally:
 - located spatially on the analyzed samples in zones whose dimensions are typically in the range 1-10 μm ,
 - depending on the depth to which the sample is analyzed.

Thus, the same object (in fact, a piece of hydrogenated metal) will present broad variations of ICs according to the place where will have been taken the sample to analyze.

IV – Short recall of AICs met apart from CHMNR experiments

Many disciplines are confronted with unexplained AICs:

- Physics of the atmosphere (ex: enrichment in ^{18}O of ozone - reaching 50 % -, of CO , CO_2 , N_2O ...),
- Cosmochemistry
 - Meteorites (many elements in various classes, e.g.: $\delta\text{D} = 5.7$),
 - Comets (e.g.: ratio D/H of Hale-Bopp 20 times the terrestrial ratio),
 - Interstellar dust (e.g.: $\delta\text{D} = 25$),
 - Molecular clouds (e.g.: $\delta\text{D} = 60$),
 - ...
- Volcanology (e.g.: ratio $^3\text{He}/^4\text{He}$ reaching 37 times the value of the atmosphere ratio),
- Geology (e.g.: ^{40}Ar excess),
- Archaeology (many anomalies of ^{14}C dating),
- ...,

It would have been interesting to examine in detail these results. But the field of study would have been too vast. This is why we will restrict ourselves to the results AICs even more problematic: those obtained for the same object.

V – Results of AICs obtained in a same sample or a same structure

Many variations of ICs on the same object, or the same structure, are measured.

As we mentioned earlier (see paragraph III), such variations are usually measured on samples resulting from CHMNRs experiments.

But they are also measured in all the disciplines meeting AICs and mentioned in the preceding paragraph.

Let us quote :

- ^{18}O of atmospheric ozone [7],
- D in the coma of the comet Hale-Bopp [8],
- many elements in different meteorites (cf infra),
- various elements in various samples of interstellar dust (cf infra),
- ^{40}Ar in terrestrial rocks (Argon datings), example: 11 and 18 My for a rock at Simplon [9],
- ^{14}C on ground (Carbon datings) (cf infra):
 - bone of a skeleton of Egyptian mummy: 2 000 and 4 000 years,
 - carrot of the Bering Strait: 6 measurements from 4 000 to 16 000 years,
 - musk ox carcass: 17 000 and 24 000 years,
 - prehistoric parietal paintings: 14 000 and 28 000 years, 15 000 and 30 000 years...
- various elements in various enriched samples for the vast 1991 IAEA Inter Comparison (cf infra).

We will examine in detail only the results obtained for 3 categories of objects: prehistoric parietal paintings, meteorites and samples of the IAEA 1991 Inter Comparison.

V.1 - Meteorites

The meteorites coming from the belt of asteroids between Mars and Jupiter, in particular chondrites which did not encounter fusion after their accretion, are carefully studied because they are a testimony of the beginning of the solar system.

Most primitive, and thus most famous, are carbonaceous chondrites, and among them C1Orgueil and ALLENDE.

It is the scientific field in which the greatest number of measurements of IC was carried out since those started nearly 50 years ago.

It is indeed Boato who, in 1954, showed the AIC of Hydrogen (Deuterium excess) in carbonaceous chondrites [10].

Then D. Black discovered in 1969 the AIC of Ne (Ne^{22} excess) in carbonaceous chondrites. In 1972, Black concluded that the origin from this Ne^{22} was an independent source, outside our solar system and then calls this excess of Ne^{22} "Ne-E", E for "Exterior" [11].

Then R.N. Clayton [12] discovered in 1973 the AIC of Oxygen not obeying the law, however well checked, of "mass fractionation" in an inclusion rich in Al, Ca and Ti of ALLENDE. He concluded from it that the explosion of a supernova (SN) near the solar system in formation injected Oxygen having its own isotopic signature in the proto-solar nebula.

Since, in inclusions called CAI - FUN for Calcium and Aluminium-rich Inclusions - Fractionation and Unknown Nuclear effects (representing however only one small part of inclusions CAI) and in the chondres (molten silicate droplets solidified very quickly), many other AICs were discovered for nearly about thirty elements: Li, B, C, N, Mg, Si, K, Ca, Cr, Ni, Kr, Sr, Zr, Mo, Ru, Pd, Ag, Xe, Ba, Sm, Nd, Sm and W.

ICs can thus vary inside the same meteorite:

- of an inclusion CAI - FUN or from one chondrite to another, distant of ten μm , even 1 μm (see for example [13]),

- of a carbonate grain to the other, grain to which the size is close to 10 μm (see for example [14]).

This type of results is not confined to meteorites. It is also obtained for interstellar dust (see for example [15]). What gives a universal character to him.

If one preserves the assumption of Clayton, i.e. SN having different isotopic signatures, it is necessary to consider today several tens of SN explosions.

Moreover, in situ decay of certain nuclides ($^{26}\text{Al} \Rightarrow ^{26}\text{Mg}$, $^{53}\text{Mn} \Rightarrow ^{53}\text{Cr}$...) place on these explosions a constraint of extremely tight temporal proximity of a few million years (see for example [16]).

Lastly, some isotopic ratios (such as $^{48}\text{Ca}/^{46}\text{Ca}$, $^{48}\text{Ca}/^{50}\text{Ti}$, $^{48}\text{Ca}/^{54}\text{Cr}$...) cannot be reproduced by calculations of stellar nucleosynthesis, some experts qualifying the situation of "intolerable" (see for example [17]).

The succession of all these discoveries thus returned gradually this type of explanation less and less probable.

With philosophy, and humour, the cosmochemists, waiting for a better understanding of these results, decided to baptize them of "FUN" for "Fractionation and Unknown Nuclear effects".

It is this term of "Unknown Nuclear Effects" (UNEs) which we chose to retain.

Because it is exactly with what CHMNRs researchers are confronted.

This is why it seems to us rational to make the bringing together between these 2 types of results.

It is also why we wish to suggest to the cosmochemists who carry out laboratory experiments to try to reproduce the AICs that they measure, to take into account the configuration of CHMNRs experiments, configuration which systematically involves Hydrogen (protium or deuterium) penetrating a metal.

V.2 - Prehistoric Parietal Paintings

The ^{14}C dating technique, conceived and used since 1948 by Libby Willard, is currently largely used.

Well adapted to periods going up to 40 000 years, this technique has consequently been very much used in the field of prehistoric cave datings, and this for several decades.

The current precision of measurements is excellent: ± 50 years at 10 000 years, ± 200 years at 20 000 years and ± 400 years at 30 000 years.

Initially a technique of counting of beta decays, the arrival in the 1980s of the new technique of Accelerator Mass Spectrometry (AMS) made possible to decrease the sample size by a factor 1 000 (today, with the new devices, the factor reaches 10 000).

This low size of samples encouraged the archaeologists to require many datings insofar as the objects were not degraded any more by the sampling. Thus most archaeologists make the same object dated several times.

Generally, measurements are coherent between them. For example, 27 measurements of samples taken on 9 paintings of bison located in the Spanish caves of Covaciella, Altamira and El Castillo: from 13 000 to 14 500 LP [18].

But sometimes, they are not, and it is what makes these values interesting for our matter.

It is indeed frequent to have variations of 5 000, 10 000 even 15 000 years:

- 6 000 years of variation for the female mégaceros of Cougnac (25 120 and 19 500 LP) [19],
- 9 000 years of variation for a horse of Chauvet (29 670 and 20 790 LP) [20],
- 15 000 years of variation for the black spots of Candamo (31 000 and 15 000 LP) [21],
-

Let us recall that a 15 000 year variation corresponds to a factor 6 on the measured quantity of ^{14}C nuclides (period 5 730 years).

However the measuring accuracy is several orders of magnitude below such a factor.

The anomaly is thus certain.

The main 3 assumptions selected to explain these variations for the same object are:

- pollution by some more recent carbon (principal assumption),
- use by the artist of charcoal more or less old,
- later final improvement of work by another artist.

These assumptions are reasonable.

However, a doubt remains when one considers the differences between various objects having a strong probability of being contemporary:

- paintings spatially close in the cave and identical in terms of subject, style and execution by the artist, present sometimes variations exceeding 10 000 years.
- paintings and other datable objects (furniture, remainders of animals, charcoals...) of the cave present sometimes variations exceeding 10 000 years.
- identical paintings in terms of subject, style and execution by the artist, present sometimes variations exceeding 10 000 years from one cave to another one.

By analogy with the results obtained for the celestial bodies and materials of CHMNRs experiments, we think that a 4th assumption, verifiable, is worth to be examined: that of the presence of in situ occurring UNEs.

Indeed, unlike the cosmochemists, the specialists in radiocarbon dating do not make the assumption of UNEs. Perhaps because they are interested only in the 3 isotopes of one element, carbon (one indeed also measures ^{13}C in order to make an isotopic fractionation correction to the $^{14}\text{C}/^{12}\text{C}$ measured proportion).

But if UNEs occurred in the pigments of these paintings, they do not have any reason to be confined with the Carbon element: measurements of IC of other elements (like Fe, Mn, Si, ...) could thus bring a valuable information.

The archaeologists could be also confronted with a problem of ICA, problem broader than a dating problem.

This is why these measurements of ICs of various elements appear to us essential.

(NB: If it proved that the AICs are measured for these elements, it would be necessary moreover to determine if these anomalies occur in zones of size 1-10 μm , as in cosmochemistry and CHMNRs experiments.)

V.3 - Samples of inter comparison IAEA 1991

This inter comparison on a large scale was carried out from 1988 to 1990, the results being published in 1991 [22].

Measurements of ICs were made in 40 laboratories of 20 countries for 13 samples especially prepared for the occasion, and enriched in D (2 samples), ^{13}C (4 sam.), ^{15}N (5 sam.) and ^{18}O (2 sam.).

The devices used were mainly Mass Spectrometers (of routine, with quadripole, with chemical ionization...) and Emission Spectrometers. A minimum of 3 determinations per isotope was required, with 6 determinations preferably, to determine the standard deviation of the average given by each laboratory.

Minimal and maximal averages obtained by laboratory:

Isotope	Echantillon	Mini (ppm)	Sigma (%)	Maxi (ppm)	Sigma (%)	Différence (%)
C13	309 A	82,22	1,6	133,60	2	63
N15	310 A	43,34	0,5	66,77	0,6	54
O18	304 B	381,39	0,2	512,33	0,1	34
N15	311	1,64	0,6	2,12	0,4	29
C13	303 A	88,11	0,5	107,50	1,3	22
N15	305 A	37,84	1,1	41,24	0,6	9
C13	303 B	439,32	0,1	477,17	0,2	8,6
N15	305 B	362,35	0,2	388,93	0,6	7,3
O18	304 A	242,95	0,0	258,24	0,4	6,3
N15	310 B	235,07	0,4	247,45	0,0	5,3
C13	309 B	524,42	0,2	544,85	0,1	3,9
D	302 B	981,39	0,5	1 015,60	0,8	3,5
D	302 A	501,25	0,3	512,53	0,1	2,3

NB: 2 laboratories obtained a null dispersion on their average.

Analyze

For the sample 304 B (water), the averages by laboratory are obtained with a sigma of about 0.1-0.2 %. But one has a difference in 34 % between laboratories. I.e 200 times more than dispersion on the average of a laboratory (the limits of detection of the instrumentation of these laboratories are respectively 0.1 and 0.3 ppm for measured values being located around 400 ppm, i.e. 2 000 times more).

For sample 310 A (urea), the difference reaches 100 times the dispersion by laboratory. For sample 311 (sulphate of ammonia), there is a factor 60 and for sample 309 A (UL glucose), a factor 35.

The smallest variation of 2.3 % recorded for sample 302 A is more than 10 times the dispersion on the average by laboratory.

If one takes the extreme of minima and maximum values (which we have eliminated from the preceding table) for which dispersion by laboratory is worse, figures are still worse.

For example, for sample 309 a: Minimal 38.70 ppm, Maximal 133.60 ppm, i.e. a 245 % difference...

These figures are thus quite simply unexplainable.

And one can only be astonished to note that the writers of the final report/ratio did not make any comment on this subject.

It would obviously be necessary to redo this type of very broad intercomparison for a better understanding of where are the most significant differences (between various types of apparatus or apparatuses of the same type).

VI – in situ UNEs and/or UNEs during measurement

Measurements appear, currently, as being reliable and precise.

However, they can give different results for the same object.

That it is for objects having a history amounting of billion years (meteorites), of tens of thousands of years (parietal paintings prehistoric) or in years (samples IAEA).

If UNEs are well the phenomenon in charge for this fact (which other one ?), then those UNEs can take place:

- during the evolution of the object itself,
- during the analysis of the sample by each laboratory,
- during both.

The 1st assumption is viable, the CHMNRs experiments are there to show it.

It will be simply noted that if AICs are quasi systematically measured during CHMNRs experiments, it is not the case for certain meteorites and certain prehistoric parietal paintings with normal ICs, the conditions to start in situ UNEs in these objects being probably not met, conditions which are always badly defined apart from the necessary presence of Hydrogen.

But taking into account of the 1st assumption involves logically the examination of the 2nd and 3rd assumptions.

The CHMNRs experiments indeed show us that UNEs occur even in media extremely close to ambient conditions, with low volumic energy density. Let us quote, among others, the experiments described in [23], [24] (70 °C, 1 Hydrogen bar), in [25] (20 °C, 6 bars of Hydrogen) and in [26] (20 °C, from 1 to 10 Hydrogen bars).

It is thus natural to raise the following question: the devices measuring ICs put the nuclides to analyze in media of energy density largely higher than that of the media mentioned above (for example the plasma of an Inductive Coupling Plasma - Mass Spectrometer, ICP-MS, the ionic bombardment of Secondary Ion MS, SIMS...). Why such media wouldn't also be the seat of CHMNRs, thus distorting the results of a class of devices compared to another one but also from one device to another one in the same class [27]?

There is also a potential methodology problem when physicists running CHMNRs experiments under very "soft" conditions, close to ambient conditions, subject then the nuclides to be analyzed in media "much more disturbed"? We think that a reflexion must be carried out in this direction.

A reflexion which should interest all the physicists thinking of the problem of the interaction experimenter - experiment, a well-known problem in Quantum Physics.

VII – Conclusions

Physicists of the atmosphere, cosmochemists, volcanologists, geologists, archaeologists all..., are confronted with problems of AICs.

In fact, the only researchers not to be confronted are :

- researchers of the disciplines using NSIC as tracers (glaciology, ecophysiology, ecology...),
- researchers in nuclear physics who proceed themselves to their nucleosynthesis in their reactors or accelerators and for which it does not exist any more reference IC,
- and, obviously, researchers who do not measure ICs.

The recent improvements and the recent wide diffusion of devices able to measure ICs explain why this problem did not appear earlier in all its width.

It is possible that UNEs being held in situ, as already the cosmochemists recognize it, are responsible for this situation.

It is also possible that certain means of isotopic analysis are themselves generating UNEs, thus distorting the results.

Thus, the author of this publication will achieve his goal if he manages to challenge:

- the cosmochemists who should think their laboratory experiments in terms of CHMNRs experiments,
- the archaeologists who should ask for the measurements of ICs in several elements in prehistoric parietal paintings,
- the experts in ICs measurements which should wonder about the non-disturbing aspect of their measurements and multiply the inter-comparisons,
- and more largely specialists in other disciplines (physics of the atmosphere, geology, volcanology...) which should take into account the potential existence of UNEs in their reflexions.

Anyway, even if these interpellations initially do not find any echo, one thing is certain: from now on, each year brings its batch of contribution in terms of experimental evidence that Unknown Nuclear Effects lead to Abnormal Isotopic Compositions.

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