Università Politecnica delle Marche
Scuola di Dottorato di Ricerca in Scienze dell'Ingegneria
Corso di Dottorato in Ingegneria Industriale

Experimental study of the possible transformation of radioactive nuclei into stable ones by ultrasound and cavitation according to the Deformed Space-Time (DST) theory

Ph.D. Dissertation of:

Andrea Petrucci

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Dipartimento di Scienze e Ingegneria della Materia, dell'Ambiente ed Urbanistica
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Ai miei genitori Giovanna e Alberto
che mi hanno sempre insegnato la bellezza e la meraviglia davanti all'ignoto e all'infinito
e che mi hanno dato la meravigliosa opportunità di perseguirli

To my parents Giovanna and Alberto,
who have always taught me beauty and wonder in the face of the unknown and the infinite
and who have given me the marvellous opportunity to pursue them.

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Abstract

The study carried on in this PhD thesis is devoted to two main targets: to study the new phenomenological theory known as Deformed Space-Time theory and to address the critical problem of what to do with the nuclear radioactive waste constantly produced by power plants. The DST theory is phenomenological and studies the four known fundamental interactions from a geometrical perspective by deforming the geometry described by the Minkowskian metric tensor which is so far the local space-time base of the four interactions.

The Minkowskian metric tensor is deformed by replacing the 4 diagonal constants by parameters that depend on the energy of the phenomenon under investigation. This theory introduces two new concepts in the dynamical description of physical phenomena: the deformed space-time for every interaction and a maximal causal speed which is finite but not limited, which depends on the energy of the phenomenon under investigation and therefore is different for every interaction. By these new concepts the DST theory is able to explain known effects that lack physical explanation, especially those in which locality and causality are missing, and is also able to predict the existence of new phenomena which are completely outside the scope of the nowadays accepted theories and therefore considered impossible according to them.

One of these new phenomena is related to the hadronic interaction for which the DST theory predicts the existence of new types of nuclear reactions. The microscopic hadronic space-time is deformed and by this deformation it takes part in the dynamics of the whole process allowing energy exchanges and processes that otherwise would be impossible. Specifically this theory predicts the existence of nuclear transmutations that increase or decrease the mass of the involved nuclei by mechanisms that have nothing to do with those known nowadays, like radioactivity, nuclear fission and nuclear fusion. In particular, the hadronic space-time deformation, once created, activates a nuclear dynamics in which the Coulomb barrier from outside the nucleus and the attractive hadronic (strong) force from inside it do not play any role in the dynamics as long as the hadronic space-time deformation is active. This space-time deformation turns out to be generated by macroscopic mechanisms that are capable of concentrating into a small space and a very short time a quantity of energy higher than the value 367.5 GeV, that is predicted by the DST theory as the energy threshold above which the hadronic space-time becomes deformed. These macroscopic mechanisms can be produced by ultrasound and cavitation in liquids or by mechanical presses applied to solid samples in which cyclic stress is produced. This PhD work was dedicated to apply ultrasound and cavitation to water solutions containing the radioactive nuclei of ^{63}Ni. The purpose of the experiments was to corroborate the predictions of the DST theory by treating the radioactive nuclei via the deformed hadronic space-time in order to transform them into stable ones and hence reduce the activity of the solutions more quickly and less dangerously than it would happen through the natural radioactive decay. The radioactive solutions were analysed before cavitation and after it by three techniques: Bremsstrahlung X-ray spectroscopy, liquid scintillation and mass spectroscopy. The results of these three techniques turned out to be compatible with each other and in favour of a consistent reduction of the activity only within 600 seconds of cavitation rather than several decades.

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Chapter 1.

The problem of nuclear wastes: their treatment and confinement nowadays

Although the nuclear reactors of new generation are equipped with various active and passive safety systems and have become extremely safe in this sense, the management of the overall process of production of nuclear energy still presents today the serious problem of dealing with the radioactive waste that is constantly produced. An identical problem, even if of lesser impact, is that concerning the treatment of radioactive waste produced in all other activities, other than power reactors, in which nuclear reactions are involved, as, for example, in medicine for the diagnosis and the treatment neoplasms. The goal of this research is precisely to address this problem and to do it in a completely different way from how it is faced today.

It is known that nowadays the great majority of radioactive waste, in particular those with a long or very long mean life time, are treated by methods that separate the radioactive part from the rest of the inert materials. This radioactive part can then be treated with ADS (Accelerator Driven Subcritical reactor) sub-critical reactors in such a way as to induce the fission of these very long-lived radionuclides and produce radionuclides with higher activity but with a significantly shorter mean life. These radionuclides are then incorporated into large volumes of inert material in order to decrease the activity per unit volume. This inert material containing the radionuclides of nuclear waste is then sealed in metal and concrete drums which are then stored in sites, possibly underground, whose geological stability guarantees the maintenance of safety conditions for time intervals of at least one hundred years, before having to intervene again to check the tightness and possibly renew the containment materials.

Chapter 2.

Do other possibilities exist? The DST suggestion

The methods briefly presented above deal with nuclear waste by known nuclear reactions (fission), by dilution of the radioactive atoms and by the confinement in underground deposits. The problem of radioactivity in this sense is not solved but only made less critical by diluting the wastes.

Conversely, a different method is proposed in this thesis. It rests on three years of my research and takes into account a new type of nuclear reactions that are studied since more than thirty years. They are identified either as LENR (Low Energy Nuclear Reactions) [1], or CMNS (Condensed Matter Nuclear Science) [1] or DST (Deformed Space-Time) Nuclear Reactions [3]. The results of this field of research are now widely verified and supported by concordant experimental evidences, obtained by different research groups working in different parts of the world, with very different experimental equipment and techniques. The nuclear reactions of this new type generally involve elements that are not considered fissionable, and therefore cannot be classified in the known nuclear fission reactions. The elements present in the experiments are usually Iron, Palladium, Nickel, Platinum, Aluminium, Titanium [1], but there is almost always the presence of Hydrogen and/or Deuterium [1]. In fact, the initial idea from which this research began, consisted in accumulating large quantities of deuterium in the crystalline lattices of these metals in order to locally lead to a very high pressure growth and hence to trigger deuterium-deuterium nuclear fusion reactions (not thermonuclear reactions). Despite the fact that this line of research is still active, over the years the experimental attention has also shifted towards other evidences, namely the constant presence, at the points where signs of reactivity is manifested either of elements not present in the composition of the materials at the beginning of the experiments, or of a different concentration (increase or decrease) of the elements initially present. In literature his evidence is referred to as transmutations and is obviously clearly nuclear in nature [5-11]. While the first two names (LENR and CMNS) of these investigations only indicate the characteristics of the reactions involved, the third name, DST nuclear reaction, calls into question a possible primary cause of these reactions, the deformation (local deformation, i.e. around where the reaction takes place) of the space-time geometry. In fact the Deformed Space-Time Theory [3,4] is able to give rather precise indications on the local conditions (i.e. at the point where the reactions have to be triggered) to trigger this new type of nuclear reactions. Furthermore, this theory states that the candidate nuclides for this new type of reaction are potentially all, whether stable or radioactive, also including the metals mentioned above. A further indication provided by the theory is that, since the energy involved in the reactions is stored in the deformation of space-time, these new nuclear reactions do not give rise to the gamma de-excitation emissions that are almost always present in the known nuclear reactions [3].

Chapter 3.

Brief overview of the Deformed Space-Time theory

When one thinks about a physical phenomenon, which could be as simple to imagine as a person walking in the street or a bird flying or pouring water from a bottle into a glass, one is accustomed to picture it in his/her mind as if that phenomenon takes place in space and in time. Or, in looking at the world around us, one is used to imagine that there is space among the objects, like the space between the posts of a soccer goal through which a football can be propelled. In this sense, space and time are the stage where physical phenomena take place. A stage in a theatre is there also when no actor is on it (strutting and fretting his hour). It has its own physical existence. Exactly in the same way space and time, to which we are accustomed, do have their own existence and in this sense they are absolute, that is they do not depend on the objects that live in them and do not depend on who is observing the objects. For every day life, this vision of a physical phenomenon is almost always correct and it was considered correct also by the physicists until the nineteenth century. Space and time were always seen as homogeneous and space as isotropic also. This simple and idyllic picture became rather shaky with the discover of electromagnetism (XIX century) which did not appear to match with the idea of absolute space-time that then had to be abandoned and considered only an approximation. The supersession of these good but incomplete concepts, formalised in the Galilean theory of relativity, with new broader ones took place with the consolidation of the new electromagnetic phenomena and the new relativity theory by Einstein, the Special Theory of Relativity [12]. In this theory, space and time became relative concepts, relative to the observer that measures them and thus began to lose their primitive almost metaphysical nature. Anyway they conserved their homogeneity and isotropy. This incredible broadening of the concepts of space, time and relativity which was achieved after about three centuries of use of the Galilean relativity appeared soon to have problems in describing phenomena either where the gravitational force played a dynamical role or involving space-time references which possessed an acceleration, i.e. are not inertial. In these systems of reference an object appears to be subjected to forces coming out of nowhere, the so called inertial forces that exist because objects possess an inertial mass. In trying to figure out how to adapt special relativity to the non-inertial frames, Einstein discovered that there was an equivalence between gravitation and accelerated references which can be based on the numerical equality of the gravitational mass and inertial mass. This was the inception of the General Theory of Relativity. Einstein found out that, apart from the generalisation of the relativity principle, his ideas were leading him to a complete re-examination of the concept of space-time. Also thanks to the mathematical achievements of Bernhard Riemann on the differential geometry of a curved multidimensional space, Einstein managed to show that the geometry of space-time on a global scale is neither flat, nor isotropic, nor homogeneous and that this geometry (distances and angles) is generated by any form of energy (mass as well, of course). In other words, space and time do not exist a priori and in this sense do not possess a predetermined flat geometry (homogeneous and isotropic) where physical objects move and live, but rather the objects with their energy and their mass generate their own curved space-time. This statement may sound as if the general relativity theory might be already connected to the Deformed Space-Time theory (and to the ideas that energy generates its own space-time,), but, even if it is indeed quite close to it, it has several hidden theoretical and mathematical implications that make it still to rigid. In fact, the curvature of space-time is only global and only gravitational. Let us now give some clues about what the Deformed Space-Time theory is about.

3.1. Deformed Space-Time theory and the Finzi principle

In the book *"Fifty Years of Relativity" published in* 1955 [13] the Italian mathematician Bruno Finzi, stated his *"Principle of Solidarity"*, whose sentences concerning our purpose can be so translated: "*It's (indeed) necessary to consider space-time TO BE SOLIDLY CONNECTED with the physical phenomena occurring in it, so that its features and its very nature do change with the features and the nature of those. In this way not only space-time properties affect phenomena, but reciprocally phenomena do affect space-time properties."* Moreover, referring to a generic N-dimensional space: *" It can,* a priori, *be pseudo euclidean, Riemannian, non-Riemannian. But (*he wonders) *how is indeed the space-time where physical phenomena take place? Pseudo euclidean, Riemannian, non-Riemannian, according to their nature, as requested by the principle of solidarity between space-time and phenomena occurring in it."* Finzi's main purpose was to apply such a principle to Einstein's Theory of General Relativity, namely to the class of gravitational phenomena.

However, its formulation is as general as possible, so to apply in principle to all the known physical interactions. Therefore, Finzi's PS is at the very ground of any attempt at geometrising physics, i.e. describing physical forces in terms of the geometrical structure of space-time.

3.2. Special Relativity from an axiomatic point of view

Einstein based his special theory of relativity on two widely corroborated evidences of physics: the constancy and invariance of the speed of light and the principle of relativity with its incredible heuristic power. Actually at the end of the XIX century, these two evidences seemed irretrievably incompatible, but he showed that by postulating them one could find out new laws of transformation of coordinates more general than the Galilean ones that reconciled the invariance of the light speed and the relativity principle. Not arguing with this incredible achievement, nonetheless more than one hundred years later one can notice that the postulate of the constancy and invariance of the speed of light framed too rigidly the physical knowledge and above all its development. In fact the special theory of relativity was tailored on electromagnetic phenomena and hence was suited for the electromagnetic interaction but unfortunately it was and has been treated so far a priori (and not inductively as Einstein for his theory) as the theory with which all physical phenomena should comply (but neither leptonic nor hadronic interactions, nor superluminal phenomena were known at that time). However it is possible to start from two more mathematical axioms [4] which leave much more freedom to the physical phenomena and obtain exactly the same results of the Special Theory of Relativity with a broader scope. The two axioms are:

1 - space-time properties: space and time are homogeneous and space is isotropic.
2 - Principle of Relativity: All physical laws must be covariant when passing from an inertial reference frame K to another frame K0, moving with constant velocity relative to K.

Nothing is said about the speed of light and its constancy and invariance, but from these two axioms one can obtain the more general result of the existence of an invariant quantity which has the dimensions of the square of a velocity. This means of course that no restrictions exist either on the class of phenomena that comply with it or on the value of this velocity. If one refers to the Galilean theory of relativity the class of phenomena includes all those of classical mechanics and the value of the invariant speed is of course infinite, if instead one refers to the Einsteinian theory of relativity the phenomena are all those of relativistic mechanics and electromagnetism and the invariant speed is the speed of light. Therefore there is, a priori, an invariant speed for every interaction, namely, a maximal causal speed for every interaction.

One important piece of the picture, which is hidden in the sentences above, has to be made explicit. When one moves from the classical mechanical laws to those of electromagnetism and relativistic mechanics, not only the maximal causal speed changes but also the geometry of the space which is euclidean for the first and Minkowskian for the second[1]. The attempt at including the class of nuclear and subnuclear phenomena in the total class of phenomena for which Special Relativity holds true is therefore expected to imply a generalization of the Minkowski metric, analogously to the generalization from the Euclidean to the Minkowski metric in going from mechanics to electrodynamics. Indeed, the generalized metric must be equipped with a dynamic character and be not only a consequence, but also an effective description of (the interaction involved in) the class of phenomena considered. This allows one in this way to get a feedback between interactions and space-time structure, already accomplished for gravitation in General Relativity. This complies with the "Principle of Solidarity".

3.3 Energy, the Finzi principle and the Deformed Space-Time Theory

At present, General Relativity (GR) is the only successful theoretical realization of geometrising an interaction (the gravitational one). As is well known, energy plays a fundamental role in GR, since the energy-momentum tensor of a given system is the very source of the gravitational field which is in turn expressed by the metric tensor that is the geometrical (metric) structure of space-time.

However this circumstance that has been so well made clear for the gravitational interaction is also present, although at a less evident level, for all the other interactions. When the problem of absolute space and time was tackled it became clear that the principle of relativity could not be formulated in terms of a coordinate system in absolute space because it lacked significance. To arrive at a precise definition it was introduced the concept of inertial frame that is a frame of reference in which the law of inertia holds in its original form. This means that the geometry of space-time is defined by the laws mechanics, which are in turn expressed in terms of a Hamiltonian, i.e. the total energy of the system, and depends on the interaction. As Max Born writes in his book, Einstein's theory of relativity [14], "It is not space that is there and that impresses its form on things, but the things and their physical laws determine space". It is then somehow possible to start from this concept and think that every process of energy exchange that takes place at the maximum causal speed characteristic of the give interaction realises the solidarity principle of Finzi since these exchanges

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[1] For the sake of clarity but also of brevity, it is made clear only in this note that if one wants to include in the theory of relativity another class of phenomena one is forced not only to change the geometry of the space (mathematically speaking) but also to add a new coordinate as it happens in passing from classical mechanic to electromagnetism, namely from Euclidean metric (3 dimensions) to the Minkowski metric (4 dimensions).

determine the geometry of space-time and at the same time this geometry affects the propagation of the interaction. In order to realise mathematically these consideration, it is possible to start from the most basic geometry of physical phenomena, the Minkowskian metric, and deforming it by letting its 4 diagonal coefficients depend on energy [4]:

Minkowski metric tensor: $g=\mathrm{diag}(1,-1,-1,-1)$ (1)

Energy deformed Minkowski metric tensor: $\eta(E)=(b_0^2(E),-b_1^2(E),-b_2^2(E),-b_3^2(E))$ (2)

where E is to be read as the total energy E exchanged by the physical system considered during the interaction process.

An energy dependent metric of this sort assumes a dynamic role, thus providing a geometrical description of the fundamental interaction considered. The generalization of the Minkowski space implies new generalized transformation laws, which admit, as a suitable limit, the Lorentz transformations (just like Lorentz transformations represent the Galilei-Newton transformations at infinite causal speed). Then, the solidarity principle allows one to recover the basic features of the relativity theory in the Lorentz (not Einstein) view (Lorentzian relativity), namely different interactions entail different coordinate transformations and different invariant speeds.

Generalizing such an argument, we can state that exchanging energy between particles amounts to measure operationally their space-time separation.

The generalised interval for the deformed Minkowski metric becomes as follows:

$$\mathrm{ds}^2=b_0^2(E)c^2\,\mathrm{dt}^2-b_1^2(E)(\mathrm{dx}^1)^2-b_2^2(E)(\mathrm{dx}^2)^2-b_3^2(E)(\mathrm{dx}^3)^2 \qquad (3)$$

It is worth to recall that the deformation of the metric, resulting in the interval above, represents a geometrisation of a suitable space-time region (corresponding to the physical system considered) that describes, in the average, the effect of non-local interactions on a test particle. It is clear that there exist infinitely many deformations of the Minkowski space (precisely, ∞^4), corresponding to the different possible choices of the parameters b_μ, a priori different for each physical system.

Each interaction produces its own metric, formally expressed by the metric tensor $\eta(E)$ above, but realized via different choices of the set of parameters $b_\mu(E)$. Otherwise said, the $b_\mu(E)$'s are peculiar to every given interaction. The GR theory implements a geometrisation (at a global scale) of the gravitational interaction, based on its derivability from a potential and on the equivalence between the inertial mass of a body and its gravitational mass. The formalism of energy-dependent metrics allows one instead to implement a geometrisation (at a local scale) of any kind of interaction, at least on a phenomenological basis. As already stressed before, such a formalism applies, in principle, to both fundamental and phenomenological interactions, either potential (gravitational, electromagnetic) or non potential (strong, weak), local and non-local [16].

Chapter 4.

The DST theory and the nuclear interaction

In the previous chapter, a very brief overview of the DST formalism has been sketched and it has been concluded that the formalism of energy-dependent metrics allows to implement the geometrisation of any kind of interaction on a phenomenological basis. It has to be added that the coefficients of the deformed metric have to be dimensionless and that there must exist for every interaction a threshold energy E_0 to account for two things: the dimensionless coefficients that will be a function of the ratio (E/E_0) and the fact that the deformed metrics have to be connected to the Minkowski metric since they have to be able to describe the phenomena that take place also in a non-deformed space-time [4]. Every interaction has its own energy threshold which has to be determined experimentally. The way to do this is to analyse by the DST formalism the results of experiments, involving the interaction whose E_0 has to be determined, which show data at variance with the theoretical predictions (which of course are calculated by a formalism that comply with the special relativity and then with a flat Minkowskian space-time). This is exactly what was done to determine the E_0 for the hadronic interaction, $E_{0,had}$. Without giving any mathematical detail [4], the experiments which show data at variance with the theory are those involving high energy collisions. In such collisions some bosons (mesons) are emitted and they show a higher probability to be emitted within a low opening angle if they are identical rather than different. This effect is the so called Bose-Einstein correlation for boson produced in high energy collisions. The calculation made by the canonical quantum formalism of the wave functions does not produce the correct value of this probability. This problem was fixed by assuming that the space-time inside the interaction region, where the pions are produced, is deformed. Therefore the deformed metric with its parameters was used to modify the scalar products involved in the calculation.

The expressions obtained by the deformed quantum formalism, that contain the undetermined $b_{\mu,had}$ coefficients of the deformed hadronic metric, were applied to the data obtained in the CERN experiment UA1 carried out in 1984. This fitting of the experimental data by the "deformed" expressions determined the functional dependence of the coefficients $b_{\mu,had}$ on the energy E and the threshold energy $E_{0,had}$. They are reported in the following Fig.1:

$$\begin{aligned}
\eta_{\text{strong}}(E) &= \operatorname{diag}(b^2_{0,\text{strong}}(E), -b^2_{1,\text{strong}}, -b^2_{2,\text{strong}}, -b^2_{3,\text{strong}}(E)) \\
b^2_{0,\text{strong}}(E) &= b^2_{3,\text{strong}}(E) \\
&= \begin{cases} 1, & 0 \le E \le E_{0,\text{strong}} = 367.5 \pm 0.4\,\text{GeV} \\ (E/E_{0,\text{strong}})^2 & E_{0,\text{strong}} < E \end{cases} \\
b^2_{1,\text{strong}} &= (\sqrt{2}/5)^2 \\
b^2_{2,\text{strong}} &= (2/5)^2.
\end{aligned}$$

Fig. 1 Deformed metric tensor for the hadronic/strong interaction

The intervals of energy shown in the figure mean that only if the energy involved in the experiment is above 367.5 GeV the hadronic interaction becomes non Minkowskian, below this value its behaviour is in accordance with special relativity. The experiments that are going to be described in the following chapters will show the application of ultrasonic waves applied to liquid solutions in order to induce in them cavitation, that is an abrupt and violent bubble collapse. The purpose of this research is to trigger new types of nuclear reactions. They are new because they are brought about by a deformed hadronic interaction that is in a deformed space-time. These reactions will have to transform a radionuclide into stable nuclei without following its natural radioactive decay. Let's briefly consider why and how ultrasonic cavitation can induce nuclear reaction either standard or deformed [3,16]. Cavitation is an implosive collapse of a gas bubble within a liquid under suitable pressure conditions. The speed of sound in water is in the order of 10^3 m/s and if the frequency of the ultrasound is 10^4 Hz the corresponding wavelength is 10^{-1} m. The bubbles contained in a liquid at room temperature have a radius of the order of 10^{-6} m. In order to have the bubble implode symmetrically the ultrasound has to be converted into a symmetric spherical shock-wave on the bubble surface. This occurs when the wavelength is much bigger than the bubble radius, which is just the case for the numbers given above. When a bubbles collapses its content, which has a pressure orders of magnitude lower than the external, escapes from the volume of the bubble to the outside, while those atoms trapped in the surface of the bubble are forcibly pushed into a spherical space whose volume becomes smaller and smaller down to a radius that can be compatible with the nuclear radius, since the power density on the bubble surface can grow to values of 10^{46} eV/s cm-2 which corresponds, for a low-density plasma, to a temperature of 10^{20}K. This power density allows a heavy ion fusion-like process. In other words the bubble acts as an inertial accelerator of neutral atoms during its collapse. This model here-sketched does not consider several drawbacks that raise some questions. One question is for instance about the state of the products from the fusion of heavy ions. These products are produced in a very excited rotational state which would either bring the child nuclide to fission or to a de-excitation by emission of gamma photons. However, these two possible occurrences do not happen in the experiments that have been performed so far. The explanation can be easily found in the DST theory. In fact the values of the energy per unit surface that have been mentioned a few lines above correspond to an energy, provided to the atomic system that collapses with the bubble surface, greater than the hadronic threshold 367.5 GeV. This means that these processes take place in a deformed space-time which contributes to the dynamics of the process by absorbing the energy that otherwise would be emitted as gamma photons.

Chapter 5.

The initial experimental tests of the DST theory with ultrasound and cavitation on stable and radioactive nuclei

On the basis of the theoretical and phenomenological considerations that have been made in the previous chapter, several experiments of different kinds have been performed in the past years since 1998 until today. In this chapter, in order to corroborate by experimental results the concepts and ideas so far presented and to give a more solid ground to the experiments and results that will be extensively shown in the next chapters, the past experiments will be very briefly listed by giving only very few details and presenting the results without any comment. The chapter is structured in mini paragraphs entitled by a name indicating the type of experiment.

Ultrasound in deionised water

Starting from 1998, three main experiments of cavitated water were performed in Italy: two in Perugia [17, 18], the last one in Rome [19].In all of them ultrasound at 20 kHz were used to sonicate bi-distilled deionised water. In all of them ICP-MS analyses (mass spectrometers) and comparisons with non sonicated sampled showed the variation of several elements of different atomic mass.

Ultrasound in water solution of stable nuclides

Water solutions of Lithium Chloride, Aluminium Chloride, Iron Chloride and Iron Nitrate (1 ppm, in deionized and bi-distilled water) and a reference sample of pure water were sonicated at a frequency of 20 kHz. Neutron passive detectors were used to measure the emission of neutrons which were indeed emitted without the accompanying gamma rays. At the same time in a suitable separated room, detectors of the same type measured the background intensities [20].

Six further cavitations of water solutions were carried out with different concentrations of $Fe(NO_3)_3$, 1 ppm, 10 ppm and 100 ppm and different ultrasound power, 100 and 130 W. A further cavitation of $Fe(NO_3)_3$ was performed at 1000 ppm. Both passive and active neutron detectors were used which, despite their completely different technology, agreed in measuring neutron bursts (not a constant flux) [20, 21].

Brittle fracture of rocks

Since the key role to generate the DST reactions is played by pressure and by a mechanism that abruptly and violently releases it, further experiments were performed by mechanical presses applied to solid materials, both natural rocks and commercial materials. Studies of rocks subjected to fracture were performed in Torino (Italy) [22-25]. Four samples, two made of green Luserna granite (gneiss) and two of Carrara marble (calcite), were compressed at a controlled displacement rate of the piston (10 μm/s). A helium-3 detector for neutrons was suitably protected by polystyrene and placed at 10 cm from the samples. At the instant of the brittle fracture it showed neutron pulses without gamma rays. These results were obtained with bars of granite that contain iron and that are subjected to brittle fracture but not with marble which do not contain iron and whose fracture is more ductile and hence not abrupt.

Ultrasound on iron bars

Cylindrical bars with 1 cm radius and 20 cm height and made of sintered Ferrite (α-iron) or of steel were treated by ultrasound at 20 kHz [26,27]. As in all the previous experiments neutrons, captured by active detectors, were emitted in bursts after a delay time and without gamma emissions. As in the other cases the delay time indicated that a preload of energy in the material is necessary, which is connected to the existence of the energy threshold in the DST metrics

An unexpected effect, however, was the appearance of circular, macroscopic and regular damage spots on the lateral surface of all the treated samples. In the case of the carbon steel bars they were brownish and had a diameter of 2-3 mm. A semi-quantitative microanalysis of some damaged regions was performed by a Zeiss Supra 40 FESEM electron microscope equipped with an energy dispersive X-ray (EDX) detector. The results were compared with those obtained from the apparently not damaged zones. Elements lighter than Iron, which were not present in the latter, were found in the former. In addition, the decreased concentration of iron is balanced by the increased amount of carbon and oxygen atoms.

Compression of iron bars

The analyses by EDX showed some interesting variations of elements. If one tries to conjecture the possible DST reactions by trying to balance together the protons and neutrons of the elements that varied, these variations indicate a possible emission of alpha particles. In order to check this point, a study was performed on steel bars subjected to pressure. The investigated material is AISI304 steel subjected to cycles of compression and relaxation at different strain rates [28]. The investigation is based on the parallelism between bubble collapse in liquids and the collapse of the cavities contained in the bars with their content, created during the cooling part of their production process. Three ZnS detectors of alpha particles registered during the strain/stress cycles signals higher than the maximum background intensity. A further support to this conclusion came from the analysis of CR39 polycarbonate detectors for alpha particles that were suitably put in contact with the surface of the bars.

Ultrasound in mercury

In the experiments with mercury and ultrasound, the idea was to fulfil two conditions at the same time. The first one had to do with the evidence that, although DST nuclear reactions can involve any nuclide (from the lightest to the heaviest), the most strongly bound nuclides respond with the shortest time delay before the enkindling of the DST reactions. From this perspective the nuclide to use had to possess a binding energy per nucleon similar to that of Iron, about 8.8 MeV. The second condition to fulfil was that the substance to treat by ultrasound were liquid in order to facilitate cavitation. These two conditions coexist perfectly in mercury. By treating a mole of mercury by ultrasound at 20 kHz for three minute in DST conditions part of the liquid mercury was transformed into a solid material [29,30,31]. In analysing this transformed material by EDS, elements and isotopes were found which were not present in the initial non treated mercury.

Ultrasound in water solutions with radioactive elements

A 300 ml water solution containing a small amount of ^{228}Th was cavitated by ultrasound at 20 kHz for 90 minutes [32]. An equivalent quantity with the same amount of ^{228}Th was used as reference. Both alpha particle measurements by polycarbonate detectors CR39 and ICP-MS measurements of the solution were carried out. Their results agreed and both indicated the reduction of ^{228}Th after 90 minutes of treatment. Some years later another solution containing the radioactive element ^{63}Ni was subjected to the same treatment of cavitation by ultrasound at 35 kHz for 100 seconds. The radiation emitted was the Bremsstrahlung X-rays of the slowed down beta particles emitted by the radionuclide. A significant decrease of 13% of this radiation was measured by a NaI gamma spectrometer after the treatment [33]. The amount of ^{63}Ni was reduced in an interval of time much shorter than that of its natural radioactive decay.

Chapter 6.

Planning of the experimental campaigns described in this thesis

The experimental campaign, carried out in the last three years, is mainly devoted to test and study the effects of ultrasounds on radioactive nuclides and in particular the possibility that the treatment, carried out according to the prescriptions of the DST theory, could transform the radioactive nuclei into stable ones. In this sense, we performed differential measurements between values of quantities related to the concentration of the radionuclide before the ultrasonic treatment and after it. These measurements have been performed by three different techniques, two involving the detection of an ionising radiation, Liquid Scintillation (LS) and Bremsstrahlung X-ray spectrometry, one involving directly the radionuclide concentration, that is mass spectrometry (ICP-MS). Other ultrasonic treatments have also been performed on stable nuclides

6.1. The radioactive and stable nuclei to treat by ultrasound and cavitation

Several might have been the choices of interesting radionuclides to treat, like ^{60}Co and above all those that are produced by ^{235}U fission and that can be found in the waste of the nuclear power plants like ^{137}Cs, ^{85}Kr, ^{241}Am, ^{113m}Cd, ^{126}Sn, ^{93}Zr, to mention only some of them that emit also gamma rays. Unfortunately, due to the strict rules that exist to deal with unsealed radioactive material and to those regarding the radiation protection, the choice made was for a pure beta radionuclide, that is ^{63}Ni. This radionuclide has a half-life of about 98.7 years and decays by emitting an electron without the following gamma relaxation which would represent the most dangerous part for the radiation protection. The beta spectrum is continuous, as is well known, with maximum energy of 66.98 keV and medium energy of 17.43 keV [34]. This choice of ^{63}Ni was also steered by its half-life time which is long enough that one could disregard its decrease due to its natural radioactive decay during the experiments. In fact, since the target of these experiments is to give evidence of a decrease of the concentration of the radionuclide due to the DST nuclear reactions, the natural radioactive decrease via β decay had to be the lowest possible, possibly several orders of magnitude lower than the DST decrease.

With regards to the stable nuclides to treat by ultrasound and cavitation, three atomic species were chosen: iron, nickel and copper. Iron was chosen since almost every previous experiment, aimed at studying the predictions of the DST theory, was carried out with liquid solutions containing this atom. In fact, according to the theory, this atom is the most favourite to start the DST reactions in matter, being located at the highest position in the Binding Energy (BE) per nucleon curve. This choice was made to broaden the collection of experimental data with it.

Stable nickel was chosen in order to compare its behaviour against that of its radioactive isotope ^{63}Ni. Stable copper, on the contrary, was chosen since the decay product of ^{63}Ni is just the natural occurring ^{63}Cu.

6.2. The experimental equipment to measure the radioactivity and the concentration of nuclei: liquid scintillation(LS), Bremsstrahlung X-ray spectrometry, mass spectrometry (ICP-MS)

The chosen radionuclide, ^{63}Ni, is a pure beta emitter whose leptonic decay is:

$$^{63}Ni \rightarrow {}^{63}Cu + e^{-} + \overline{\nu}_{e}.$$

The concentration of ^{63}Ni contained in the solutions treated by ultrasound can be measured by three different types of techniques. The first one is a radioactive technique that exploits the kinetic energy of the emitted electrons, the beta particles. This energy can be detected by a liquid scintillation detector (LS) in which the electron strikes the organic molecules of the liquid scintillator and brings about the emission of light pulses from them. The photons of the light pulses travel through the liquid solution without high attenuation and are collected by photomultiplier tubes that surround the radioactive sample. The liquid scintillation instrument, that was used to detect the emitted electrons and hence to measure their counts per second before the ultrasonic treatment and after it, is the Hidex 300 SL [35], shown in Fig.2.

Fig.2. Liquid scintillation counter used to measure the count rate of ^{63}Ni in the solutions before and after the ultrasound treatment.

In order to carry out these measurements, some tiny quantities (micro-litres) of the radioactive solution, (either blank, i.e. non-treated by ultrasound, or sonicated) have to be sampled from it, then chemically treated to reach a suitable acidity and finally mixed with the organic liquid scintillator. In particular the scintillation counter is fed with 8 ml of radioactive solution, whose pH has been raised to a value between 1.4 and 2, mixed 12 ml of liquid scintillator.

This instrument is capable of detecting very low levels of radioactivity. More specifically it possesses a Minimum Detectable Activity (MDA) of 0.017 Bq/ml for Tritium in water with a counting time of 1 hour. This feature played an important role in these experiments since it was decided to carry out tests with ^{63}Ni both with high and low activity [36].

There exists a second possibility to measure indirectly the radioactivity of ^{63}Ni. As is well known when an electric charge varies its velocity, in other words it is accelerated or decelerated, it emits an electromagnetic field whose energy is higher the higher the acceleration is. This is what happens to the electrons emitted by ^{63}Ni. Their kinetic energy is dissipated as they move through the solution and the glass walls of the vessel and interact with the ions and atoms, it is the phenomenon known as Bremsstrahlung (braking radiation). In this case the electromagnetic energy emitted lies in the range of hard X-rays and its spectrum is not quantised. Theoretically this energy varies from a few keV to the maximum energy of the emitted electrons that is 66.98 keV. In other to detect and count the X-ray photons in this range, two solid scintillating detectors of sodium iodide (NaI) were used: the Atomtex AT6102 A which contains a cylindric crystal of NaI 40 X 40 mm and an Harshow cylindric 3x3 inches NaI crystal. The first one was already equipped with its own circuitry (high voltage supply, pre-amplifier, amplifier, MCA) and its own display. The second one had its own photomultiplier and was connected to an Ortec digiBASE which contains the high voltage supply, the pre-amplifier, the amplifier and the MCA and can be connected to a computer via USB to visualise the spectrum.

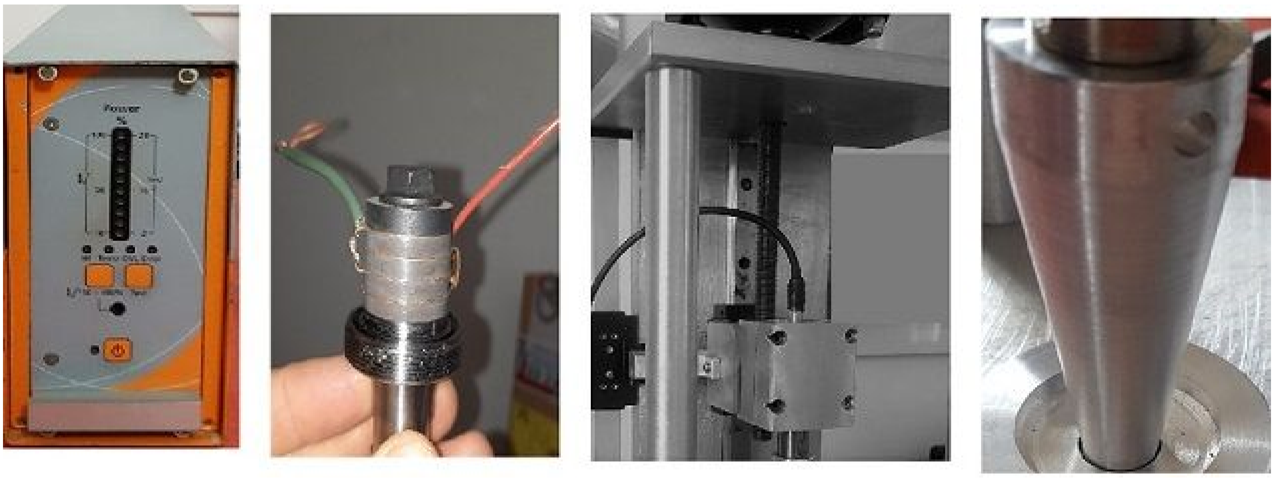
The NaI spectrometers are usually inserted in a well made of bricks and plates of lead and copper to shield radiation. Thus the X-ray and gamma ray background is cut down in order to increase the resolution of the spectrometers. However in these experiments, the ultrasound machine by which the radioactive solutions were sonicated was too large to fit into the shielding wells. This circumstance meant to make a choice between two possibilities: either to move the vessel containing the radioactive solution several times back and forth from the ultrasound machine to the NaI spectrometers placed in the shielding well, or to use the NaI spectrometers without shielding.

The first choice means to have a poor control of the position of the vessel both in the ultrasound machine and in the well near the spectrometers. Of course, this implies a large uncertainty of the geometrical detection efficiency, but at the same time a lower X-ray background and a better spectrum. On the contrary, the second choice means not to move the vessel from the initial position in the ultrasound machine and to place the spectrometers outside the shielding around the vessel in a fixed position once for all, which implies a fixed geometrical detection efficiency, but a higher X-ray background and hence spectra with lower resolution. Since the activity of the available ^{63}Ni was rather high, MBq (mega Becquerel) that is much higher than the count rate of the X-ray background it was decided for the second choice.

6.3. The ultrasound machine

There are several choices of ultrasound machines because high-frequency acoustic waves have different applications both in chemical laboratories (sonicators) for cleaning, mixing, homogenizing, breaking chemical bonds and also in industry, for example for welding plastics with intense heat locally generated by ultrasound.

Generally speaking, the welding ultrasound machines used in the industry are sturdier than sonicators and ultrasonic baths and are able to transfer a much higher power to the materials to which they are applied. When the first experiments were planned [37] the ultrasonic welding machines appeared to be more suitable for our purposes which imply long treatments of different types of materials either liquid and solid. Thus the ultrasound machine that has been used in these experiments is a welding machine. The machine is basically made of three main parts: the high frequency electrical generator; the piezoelectric converter; the sonotrodes or (horns), see Fig.3.



(a) **(b)** **(c)** **(d)**

Fig. 3 Main parts of the ultrasound generator
(a) Generator of electrical oscillation, 35kHz; **(b)** converter content i.e. 3 disks of piezoelectric ceramics;
(c) converter enclosed in its case with coaxial cable that brings the oscillating electrical signal to be converted;
(d) sonotrode connected to the converter, it concentrates and conveys mechanical oscillations to the sample.

The generator DN400M is produced by the EUROSONIC Neumann GmbH Ultraschall-Technologie. It is capable of a maximum output power of 400 W at 35 kHz. The high frequency voltage (that ranges between 600 to 1000 volts) is provided to the converter that converts the electric oscillation into a mechanical one by three disks of piezoelectric ceramic. The generator has two buttons: one to turn it on and off, one to test the operation and figure out if the whole settings are correct. Besides, there is a screw that can be turned with a small screwdriver in order to set the amplitude of the oscillation which can range from 50% to 100% of the maximum. Above the buttons, there are several LEDs red or green. The red ones indicate, when they turn on, possible failures: an overload; a too high temperature of the ceramics with the risk to break them or to make them loose the piezoelectric properties; an impedance mismatch between the frequency of ultrasound with respect to the sonotrode dimensions. In fact, due to the thermal expansion, these dimensions may vary to the point that the generator's electronics cannot adapt the frequency any longer. This may result in a reflected wave travelling back to the piezoelectric ceramics which may be thus easily damaged. The green LEDs, on the contrary, are an indicator of the power transferred to the sample subjected to the ultrasound.

Chapter 7.

First campaign of measurements: tests on liquid solutions of the stable nuclei Ni and Cu

The first measurements that were planned and executed were on non-radioactive material. Although the radioactive vials of ^{63}Ni were already at disposal and it was possible to begin the preparation of the radioactive solutions, the decision was made to gain some experience of the new ultrasound equipment with stable solutions that, of course, required a lower level of care in handling them. As reported in the paragraph 7.1, the chosen stable nuclides to treat by ultrasound were nickel, copper and iron, because of their relation to the following experiments with ^{63}Ni and to the previous experimental campaigns carried out in the past years. The experimental activities will be reported and described with the same temporal order in which they were actually performed. In this sense, in this chapter only the experiments carried out on nickel solutions with concentration of 1, 5 and 10 ppm and on copper solutions with concentration of 5 and 10 ppm will be presented.

7.1. Preparation of the liquid samples to sonicate

From two Normex ampoules containing high purity solutions for AAS (Atomic Absorption Spectroscopy), one of Ni and one of Cu, two 1000 ppm standard solutions of 1 L each of Ni and Cu were prepared. The number of cavitations to be performed depended on 3 sets of discrete variables: the concentrations, 3 for nickel, 2 for copper; the duration of application of ultrasound that was decided to be 100 seconds, 200, seconds, 300 seconds; the amplitude of ultrasound that was decided to be 50% and 70% of the maximum[2]. By mixing these variables, 21 solutions of Ni had to be prepared which included those to be cavitated and the blanks, and 14 solutions of Cu. It is easy to see that there were 7 solutions of Ni at 1 ppm, 7 of Ni at 5 ppm, 7 of Ni at 10 ppm, 7 of copper at 5 ppm and 7 of copper at 10 ppm. Since the volume of solution to cavitate was 25 ml, three flasks of 200 ml were prepared at the three concentrations (1, 5, and 10 ppm) for Ni and 2 flasks of 200 ml at the two concentrations (5 and 10 ppm) were prepared for Cu. All the cavitated solution samples and the non-cavitated ones, the blanks, at different concentrations, 1, 5, 10 ppm were drawn from these 200 ml of standard solutions. This procedure guaranteed a high degree of homogeneity among all the samples and more over between a cavitated sample and its related blank. Each sample was prepared just before it had to be cavitated. The samples (Table 1 and Table 2) were produced straight away in the glass vessels where they would be treated by ultrasound soon after their preparation. Starting from the solutions at 1000 ppm all the required dilutions were made with bi-distilled deionised ultra pure water at 18.2 MΩ by pipettes and micro-pipettes in order to reach the more diluted samples.

Table 1: 21 solutions of Ni with three different concentrations 1, 5 and 10 ppm, cavitated at 50% and 70 % of the ultrasonic amplitude for 100, 200 and 300 seconds.

Ni 1 ppm blank	Ni 1 ppm 50% 100 s	Ni 1 ppm 50% 200 s	Ni 1 ppm 50% 300 s	Ni 5 ppm blank	Ni 5 ppm 50% 100 s	Ni 5 ppm 50% 200 s	Ni 5 ppm 50% 300 s	Ni 10 ppm blank	Ni 10 ppm 50% 100 s	Ni 10 ppm 50% 200 s	Ni 10 ppm 50% 300 s
	Ni 1 ppm 70% 100 s	Ni 1 ppm 70% 200 s	Ni 1 ppm 70% 300 s		Ni 5 ppm 70% 100 s	Ni 5 ppm 70% 200 s	Ni 5 ppm 70% 300 s		Ni 10 ppm 70% 100 s	Ni 10 ppm 70% 200 s	Ni 10 ppm 70% 300 s

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[2] These decisions about the duration and the amplitude of ultrasound were made on the base of several test previously performed in which it was controlled the stability of the ultrasound machine and the temperature that was reached. Usually these ultrasound welders are cooled by compressed cold air. The machine at our disposal, however, was not provided with the cooling circuit. This explains why only limited durations of application of ultrasound could be used and the amplitude could not be higher than 70%.

Table 2: 14 solutions of Cu with two different concentrations 5 and 10 ppm, cavitated at 50% and 70 % of the ultrasonic amplitude for 100, 200 and 300 seconds.

Cu 5 ppm blank	Cu 5 ppm 50% 100 s	Cu 5 ppm 50% 200 s	Cu 5 ppm 50% 300 s		Cu 10 ppm blank	Cu 10 ppm 50% 100 s	Cu 10 ppm 50% 200 s	Cu 10 ppm 50% 300 s
	Cu 5 ppm 70% 100 s	Cu 5 ppm 70% 200 s	Cu 5 ppm 70% 300 s			Cu 10 ppm 70% 100 s	Cu 10 ppm 70% 200 s	Cu 10 ppm 70% 300 s

7.2. Protocol of cavitation of the liquid samples and preparation of the samples for ICP-MS

Each sample mentioned in the tables above, but the blanks, was cavitated according the following protocol:

- measurement of the weight of the clean, dry and empty glass vessel;
- 25 ml of the standard solution added in the vessel by laboratory pipettes;
- measurement of the weight of the vessel with the solution;
- 5 ml of freshly deionised ultra pure water (18.2 MΩ) added in the vessel by laboratory pipettes;
- measurement of the weight of the vessel with the solution;
- positioning of the vessel on the stand in the ultrasound machine under the sonotrode;
- measurement of the temperature of the solution by an infrared thermometer FLUKE 69 [38];
- lowering of the sonotrode into the solution in the vessel down to a certain distance from the vessel bottom;
- switching on ultrasound;
- measurement of the duration of application and video clip recording of the cavitation;
- turning off ultrasound;
- sonotrode raised back out of the solution;
- measurement of the temperature of the solution by an infrared thermometer FLUKE 69;
- measurement of the weight of the vessel with the solution after treatment;

After each cavitation the cavitated sample was poured into a 50 ml volume Falcon tube which was air tight sealed by a Parafilm tape and completely wrapped up by aluminium foil in order to protect it from the light. The same procedure was applied to the three blank samples. All the samples were given to the Italian National Research Council for mass spectrometry analyses (ICP-MS).

Between one cavitation test and the next the vessel was washed five times with deionised ultra pure water and so was the sonotrode.

Chapter 8.

Second, third, fourth and fifth campaigns: experiments with liquid solutions of ^{63}Ni with high activity

An Amersham liquid radioactive source of ^{63}Ni with a volume of about 150 ml was made available to perform the ultrasonic tests on high activity materials. The activity of this solution at the date when it was provided was of the order of several tens of MBq. From this initial solution four samples of 30 ml each of ^{63}Ni could be produced by some dilutions with deionised ultra pure water and by adding nitric acid (HNO_3) from a high concentration standard for atomic absorption spectroscopy. By these four sample, four ultrasonic tests could be carried out. Each solution was prepared immediately before the cavitation process and directly in the glass vessel where cavitation would be induced.

8.1. Preparation of the four liquid samples to sonicate

The four solutions at high activity of ^{63}Ni were produced from the Amersham liquid radioactive source by several dilutions, as it was said. Although each of them was made in a different day (the day when that solution had to be cavitated) their features will be reported together in this paragraph. As it will be seen (Table 3), these four samples of solution of ^{63}Ni have not exactly the same values of activity and acidity. The common thing is that these two characteristics are rather high. The choice to produce samples of very high acidity, i.e. very low pH, was made because this guarantees the stability of the solute, ^{63}Ni, in the solution. The high concentration of the H_3O+ cations prevents the positive ^{63}Ni ions from forming salts with other anions in the solution and from adhering to the walls of the glass vessel or to those of the sonotrode.

Table 3: Characteristics of the four solution samples with high activity of ^{63}Ni

	First sample **24th July 2020**	**Second sample** **17th Nov 2020**	**Third sample** **10th Dec 2020**	**Fourth sample** **17th Dec 2020**
Quantity	30 ml	30 ml	30 ml	30 ml
Activity	13.2 ± 0.4 MBq	2.72 ± 0.1 MBq	2.90 ± 0.1 MBq	5.47 ± 0.2 MBq
pH [H_3O+] of HNO_3	-0.96 9.1 M	-0.76 5.75 M	-0.76 5.75 M	-0.84 6.92 M

8.2. Measurement protocol: cavitation, X-ray spectra, samplings of the solution for LS measurements

With respect to the cavitations of the solutions with stable atoms described in the previous chapter, the measurement protocol was much more complex in this case since one had to perform several radioactive measurements before, during and after cavitation and had to sample the cavitated solution in order to set aside, at certain points, some small quantities of it to prepare the samples to analyse by liquid scintillation.

The ^{63}Ni samples described in the previous paragraph were treated according the following protocol reported in Table 4:

Table 4: Sequence of operations to cavitate the radioactive samples and keep track of the temperature, mass, vessel disposition, cps of X-rays, solution samplings for LS analysis.

step	Operation	Operation explanation
1	measurement of the weight of the clean, dry and empty glass vessel;	weight of the container to subtract from the gross weights (container and solution).
2	30 ml of radioactive solution added into the vessel;	
3	measurement of the weight of the vessel with the solution;	the initial gross weight.

4	positioning of the vessel on the stand in the ultrasound machine under the sonotrode;	
5	measurement of the temperature of the solution by an infrared thermometer FLUKE 69;	initial temperature before ultrasound.
6	first sampling by a micro pipette of some tens of µl[3] of the solution to set aside for liquid scintillation;	the sampling of the non-cavitated solution for liquid scintillation (blank).
7	measurement of the Bremsstrahlung X-rays for 1000 seconds;	initial X-ray measurement; the blank to refer to and to subtract from the spectra acquired after the cavitations.
8	immersion of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel;	
9	cavitation for 100 seconds at 50% of the maximum amplitude and measurement of the Bremsstrahlung X-rays during the cavitation;	X-ray measured to control the possible increase of radioactivity due to the decrease of the ^{63}Ni half life time, $t_{1/2}$, due to DST reactions[4].
10	sonotrode raised back out of the solution;	
11	measurement of the temperature of the solution by an infrared thermometer FLUKE 69;	temperature after 100 seconds of ultrasound.
12	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to check the possible decrease of radioactivity due to 100 seconds of DST reactions. Solution and sonotrode cooling.
13	second sampling by a micro pipette of some tens of µl of the solution to set aside for liquid scintillation;	the sampling of the solution after 100 seconds of cavitation for liquid scintillation analysis.
14	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to consider the subtraction of ^{63}Ni from the solution at step 13. Solution and sonotrode cooling.
15	measurement of the temperature of the solution by an infrared thermometer FLUKE 69	temperature after 1000 + 1000 seconds of ultrasound off and before the second cavitation.
16	immersion of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel;	
17	cavitation for 200 seconds at 50% of the maximum amplitude and measurement of the Bremsstrahlung X-rays during the cavitation;	X-ray measured to control the possible increase of radioactivity due to the decrease of the ^{63}Ni half life time, $t_{1/2}$, due to DST reactions (refer to step 9 for the footnote).
18	sonotrode raised back out of the solution;	
19	measurement of the temperature of the solution by an infrared thermometer FLUKE 69;	temperature after 200 seconds of ultrasound.
20	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to check the

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[3] The cavitated solution had to be sampled during the sequence of operations in order to prepare the samples for liquid scintillation. It had been decided to have not just the samples at the beginning (before ultrasound) and at the end (after the three cavitations (100, 200, 300 s) but also in-between the three cavitations. The cavitated solutions had a very low pH, while the sample for LS must have a pH between 1.4 and 2. This is the reason why the samplings of the four solutions was only a few tens of µl (65µl for the first, 95 µl for the second, 95 µl for the third and 87µl for the fourth).

[4] It has been argued that the decrease of radioactivity after the cavitation processes is not due to the transformation of the radioactive nuclei into stable ones thanks to the DST reactions that transform them by the deformation of the space-time of the hadronic interaction, but rather it is due to the effects of the ultrasound on the half-life time of ^{63}Ni which becomes shorter. This hypothesis, confronted with the decrease of ^{63}Ni reported in the results, implies a tremendous increase of radioactivity during cavitation. Thus, by measuring the X-rays during the cavitation process, one keeps an eye on the possible increase of radioactivity due to the possible decrease of the ^{63}Ni half-life time.

		possible decrease of radioactivity due to 200 seconds of possible DST reactions. Solution and sonotrode cooling.
21	third sampling by a micro pipette of some tens of µl of the solution to set aside for liquid scintillation;	the sampling of the solution after 200 seconds of cavitation for liquid scintillation analysis.
22	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to consider the subtraction of ^{63}Ni from the solution at step 21. Cooling of the solution and of the sonotrode.
23	measurement of the temperature of the solution by an infrared thermometer FLUKE 69	temperature after 1000 + 1000 seconds of ultrasound off and before the third cavitation.
24	immersion of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel;	
25	cavitation for 300 seconds at 50% of the maximum amplitude and measurement of the Bremsstrahlung X-rays during the cavitation;	X-ray measured to control the possible increase of radioactivity due to the decrease of the ^{63}Ni half life time, $t_{1/2}$, due to DST reactions (refer to step 9 for the footnote).
26	sonotrode raised back out of the solution;	
27	measurement of the temperature of the solution by an infrared thermometer FLUKE 69;	temperature after 300 seconds of ultrasound.
28	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to check the possible decrease of radioactivity due to 300 seconds of possible DST reactions. Cooling of the solution and of the sonotrode.
29	fourth sampling by a micro pipette of some tens of µl of the solution to set aside for liquid scintillation;	the sampling for liquid scintillation after 300 seconds of cavitation.
30	measurement of the Bremsstrahlung X-rays for 1000 seconds;	X-ray measurement to consider the subtraction of ^{63}Ni from the solution at step 29. Cooling of the solution and the sonotrode.
31	vessel removed from the ultrasound machine and measurement of the weight of the vessel with the solution;	The final gross weight of the vessel and solution after the three processes of cavitation. Useful to estimate the evaporation.
32	measurement of the X-ray background	

8.3. Preparation of the blank and of the cavitated samples for LS and ICP-MS measurements

At the end of the experimental protocol several operation had still to be carried out. The four µl samples for liquid scintillation had been already set aside for further treatment. More specifically each of them had been added into a small 25 ml flask where in advance some millilitres of deionised ultra pure water add been added and where the dilutions to reach a suitable pH for the liquid scintillator would be done.
When the vessel was removed from the ultrasound machine, at step 30, the content was poured into a Falcon tube of 50 ml which was air tight sealed by Parafilm and wrapped up with aluminium foil to protect it from light. This Falcon tube together with another Falcon tube with 5 ml of the initial non-cavitated radioactive solution were respectively the cavitated and the blank samples for ICP-MS.
After the preparation of the samples for ICP-MS, those for liquid scintillation were prepared. At the steps 6, 13, 21, and 29, some tens of micro litres of the radioactive solution in the vessel were sampled and set aside in four 25 ml flasks as

already said. The content of these flasks was then brought up to volume with deionised ultra pure water and after closing them with a tight cap they were shaken in order to homogenise their content.

The containers for LS are of special type with a volume of 20 ml which has to be filled with 8 ml of the radioactive solution to analyse and 12 ml of the liquid scintillator. Several of these containers were at our disposal along with the scintillator cocktail. Thus, with the help of pipettes and micropipettes, 8 ml of the radioactive solution was sucked from each flask and added in the LS spectroscopy container. Both the ICP-MS and the LS samples were kept in the darkness and in a fridge at a temperature of about 4° C before delivering them to the laboratories where the analysis would be done.

Needless to say that after every cavitation the vessel, the sonotrode, the pipettes, the flasks and the beakers used were always rinsed five times with ultra pure water.

Chapter 9.

Sixth campaign: experiments with liquid solutions of ^{63}Ni with low activity

The low activity experiments with ^{63}Ni were performed with a Campoverde source with a calibration certificate of Eckert & Ziegler. This source had a volume of 5 ml, a total activity of 3780 Bq, pH 1 of hydrochloric acid (HCl) and a carrier concentration of 50 µg/ml.

Thanks to the high counting sensitivity of both ICP-MS and LS these 5 ml were divided into ten and thus ten different solutions each containing 378 Bq of ^{63}Ni could be used in this experimental campaign.

9.1. Preparation of the liquid samples to sonicate

Each of the 10 samplings of the standard solution was of 0.5 ml which had to be brought to a volume of 30 ml in order to be cavitated. This operation implied of course a dilution of the HCl, already present, and thus an increase of the pH which, on the contrary, had to remain low, at the highest equal to 1, in order to chemically stabilise the ^{63}Ni in the solution. For this reason a certain quantity of acid had to be added in bringing up to volume the 0.5 ml of radioactive solution. The idea was to add HCl for 5 out of the 10 samples and HNO_3 for the other 5. The dilutions were so made that the final pH of the 30 ml solution to cavitate was -0.52 for the HNO_3 and -0.5 for HCl, that is 3.35 M and 3.18 M respectively. A preliminary test with a sample with HCl acid showed that the ERGAL sonotrode was chemically attacked by the acid and somehow dissolved. Thanks to this evidence only two samples were produced with HCl, the other 8 were prepared with HNO_3 which, despite the same pH of HCl, did not damage the sonotrode.

Thus, the samples (Table 5) were prepared straight away in the cavitation vessel by adding a little quantity of deionised ultra pure water, then the 0.5 ml of the ^{63}Ni standard radioactive solution, then the right volume of HNO_3 standard for a pH 0.52 and finally by bringing up to 30 ml with other deionised ultra pure water.

Table 5: Characteristics of the 8 solution samples with low activity of ^{63}Ni

	8 Samples with HNO_3
Quantity	30 ml
Activity of ^{63}Ni	378 ± 11 Bq
Activity concentration	12.6 ± 0.4 Bq/l
pH **Molarity of HNO_3**	-0.52 3.35 M

9.2. Measurement protocol: cavitation and other measurements

With respect to the cavitations of the radioactive solutions with high activity of ^{63}Ni described in the previous chapter, the measurement protocol was simpler since several operations could avoided in this case. First of all, the Bremsstrahlung X-ray measurements were not performed since, due to the low activity of ^{63}Ni the X-ray emission that escaped outside the vessel was very low and indistinguishable from the X-ray background. Only two analyses were planned: ICP-MS and LS. Besides, for the latter, it was decided that only the non-cavitated solution and that obtained after a complete set of cavitations (100, 200, 300 seconds) would be analysed. Thus no sampling between cavitations had to be collected and set aside (see steps 6, 13, 21, 29 in Table 4 chapter 8). These low activity samples were prepared and treated according to the following protocol in Table 6:

Table 6: Sequence of operations to prepare and cavitate the radioactive samples and keep track of the temperature, mass and vessel disposition.

step	Operation	Operation explanation
1	measurement the weight of the clean, dry and empty glass vessel;	weight of the container to subtract from the gross weights (container and solution).

2	deionised ultra pure water added into the vessel	
3	precise volume of HNO_3 added into the vessel	the right quantity of acid in order to reach the wanted pH, i.e. -0.52
4	stirring of the mixture	in order to homogenise the acid and the water
5	0.5 of the ^{63}Ni radioactive solution added into the vessel	
6	solution brought up to volume, 30 ml, with deionised ultra pure water	
7	measurement of the weight of the vessel and the radioactive solution	by subtracting the value in step one from this value, the net weight of the solution is obtained
8	positioning of the vessel on the stand in the ultrasound machine	
9	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	the initial temperature of the solution
10	lowering of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel	
11	cavitation at 50% of the maximum amplitude for 100 seconds	
12	sonotrode raised back out of the solution	
13	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	temperature after 100 seconds of cavitation
14	10 minute wait with ultrasound off	cooling of the sample and the sonotrode
15	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	temperature before the second cavitation
16	lowering of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel	
17	cavitation at 50% of the maximum amplitude for 200 seconds	
18	sonotrode raised back out of the solution	
19	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	temperature after 200 seconds of cavitation
20	10 minute wait with ultrasound off	cooling of the sample and the sonotrode
21	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	temperature before the third cavitation
22	lowering of the sonotrode into the solution with its tip at a precise distance from the bottom of the vessel	
23	cavitation at 50% of the maximum amplitude for 300 seconds	
24	sonotrode raised back out of the solution	
25	measurement of the temperature of the sample with an infrared thermometer FLUKE 69	temperature after 300 seconds of cavitation
26	removal of the vessel from the ultrasound machine	
27	measurement the weight of the cavitated solution	Gross weight of the solution after the three cavitations. By subtracting the value in step 1 from this value, the net weight of the solution after cavitation is obtained. Also the quantity of the evaporated water can be obtained.

9.3. Preparation of the blank and of the cavitated samples for LS and ICP-MS measurements

At the end of the experimental protocol several operation had still to be carried out. When the vessel was removed from the ultrasound machine and its weight measured, step 18 and 19, the content was sampled to remove the right quantity (48 µl) of solution to prepare the right cavitated sample for liquid scintillation. This quantity was inserted into the LS container and was added with deionised ultra pure water up to 8 ml which is both the right volume for the LS container and has the pH suitable for LS (between 1.4 and 2, 1.7 was chosen). Twelve millilitres of scintillator cocktail was then poured into the LS container which was then capped and air tight sealed by Parafilm and wrapped up by aluminium foil to protect it from the light. The rest of the cavitated solution was poured into a Falcon tube of 50 ml which was also air tight sealed by Parafilm and wrapped up by aluminium foil to protect it from the light. With regards to the blank samples for LS and ICP-MS, one of the 8 samples prepared from the initial standard Eckert & Ziegler radioactive solution was not treated by ultrasound and was used as blank. The same preparation with 48 µl, described a few lines above, was carried out to prepare the blank sample for LS in the LS container. The rest of the non treated solution was poured into a Falcon tube also air tight sealed and wrapped up in aluminium foil to be the blank sample for ICP-MS.

Needless to say that after every cavitation the vessel, the sonotrode and all the pipettes, flasks and beaker used were always rinsed five times with ultra pure water.

Chapter 10.

Seventh campaign: tests on liquid solutions of the stable nuclei Ni, Cu and Fe

In chapter 8 the experiments of stable nuclides, Ni and Cu, were presented. In particular the solutions treated by ultrasound had concentrations 1, 5 and 10 ppm for nickel and 5 and 10 ppm for copper. Once the experiments with radioactive solutions were finished, it was decided to complete the set of experiments for Ni and Cu with the 1000 ppm solutions. Similar experiments were also performed with iron, with concentrations 10, 100 and 1000 ppm.

10.1. Preparation of the liquid samples to sonicate

It was easy to prepare the 30 ml samples in the cavitation vessel for Ni and Cu, since the 1000 ppm solutions had been already at our disposal from the previous experiments. For iron, on the contrary, the preparation of the samples began from the Normex ampoules containing a high purity solution for AAS from which 1 L of 1000 ppm solution of Fe was prepared.

The number of cavitations to be performed for the 1000 ppm solutions of Ni and Cu depended on 2 sets of discrete variables: the duration of application of ultrasound and the amplitude of ultrasound. Like for the previous solutions in chapter 8, the durations were 100, 200 and 300 seconds and the amplitude were 50% and 70%. As reported in Table 7, seven 30 ml samples of Ni and seven 30 ml samples of Cu were produced:

Table 7: 6 solutions of Ni and 6 solutions of Cu with 1000 ppm were cavitated at 50% and 70 % of the ultrasonic amplitude for 100, 200 and 300 seconds.

Ni 1000 ppm blank	Ni 1000 ppm 50% 100 s	Ni 1000 ppm 50% 200 s	Ni 1000 ppm 50% 300 s	Cu 1000 ppm blank	Cu 1000 ppm 50% 100 s	Cu 1000 ppm 50% 200 s	Cu 1000 ppm 50% 300 s
	Ni 1000 ppm 70% 100 s	Ni 1000 ppm 70% 200 s	Ni 1000 ppm 70% 300 s		Cu 1000 ppm 70% 100 s	Cu 1000 ppm 70% 200 s	Cu 1000 ppm 70% 300 s

As to the samples of iron, first of all 2 flasks of 250 ml had to be prepared with a concentration of 10 ppm and the second with a concentration of 100 ppm. Both of them were prepared from the 1000 ppm standard solution with the suitable dilution. The number of samples to prepare was 7 for each concentration, that is 21 samples as clarified by Table 8:

Table 8: 21 solutions of Fe with three different concentrations 10, 100 and 1000 ppm, cavitated at 50% and 70 % of the ultrasonic amplitude and for 100, 200 and 300 seconds.

Fe 10 ppm blank	Fe 10 ppm 50% 100 s	Fe 10 ppm 50% 200 s	Fe 10 ppm 50% 300 s	Fe 100 ppm blank	Fe 100 ppm 50% 100 s	Fe 100 ppm 50% 200 s	Fe 100 ppm 50% 300 s	Fe 1000 ppm blank	Fe 1000 ppm 50% 100 s	Fe 1000 ppm 50% 200 s	Fe 1000 ppm 50% 300 s
	Fe 10 ppm 70% 100 s	Fe 10 ppm 70% 200 s	Fe 10 ppm 70% 300 s		Fe 100 ppm 70% 100 s	Fe 100 ppm 70% 200 s	Fe 100 ppm 70% 300 s		Fe 1000 ppm 70% 100 s	Fe 1000 ppm 70% 200 s	Fe 1000 ppm 70% 300 s

10.2. Protocol of cavitation of the liquid samples and preparation of the samples for ICP-MS

Each sample mentioned in the tables above, but the blanks, was cavitated according the following protocol:

- measurement of the weight of the clean, dry and empty glass vessel;
- 30 ml of the standard solution added in the vessel by laboratory pipettes;
- measurement of the weight of the vessel with the solution;
- positioning of the vessel on the stand in the ultrasound machine under the sonotrode;
- measurement of the temperature of the solution by an infrared thermometer FLUKE 69;
- lowering of the sonotrode into the solution in the vessel down to a certain distance from the vessel bottom;
- switching on of the ultrasound;
- measurement of the duration of application (either 100s or 200s or 300s) and video clip recording of the cavitation;
- turning off of the ultrasound;
- sonotrode raised back out of the solution;
- measurement of the temperature of the solution by an infrared thermometer FLUKE 69;
- measurement of the weight of the vessel with the solution after treatment;

After each cavitation the cavitated sample was poured into a 50 ml volume Falcon tube which was air tight sealed by a Parafilm tape and completely wrapped up by aluminium foil in order to protect it from the light. The same procedure was applied to the blank samples. All the samples were given to the Italian National Research Council for mass spectrometry analyses (ICP-MS).

Between one cavitation test and the next the vessel was washed five times with deionised ultra pure water and so was the sonotrode.

Chapter 11.

Analysis of the data obtained from the solutions with high activity and stable nuclides

The radioactivity of all the radioactive samples produced, both cavitated and non-cavitated, was measured both by liquid scintillation (the radionuclide was ^{63}Ni a pure beta emitter) and by X-ray spectrometry that recorded the Bremsstrahlung emission of the beta particles as they were slowed down in the solution and in the glass of the vessel. The Bremsstrahlung X-ray spectrometry was carried out directly by the experimenter during the cavitation by two NaI detectors as it was described in chapter 7. On the contrary, the beta liquid scintillation was entrusted to a laboratory accredited for radioactivity measurements. Besides all the radioactive samples and those with stable atoms, were also delivered to a laboratory of the Italian National Research Council (CNR) for measurements by mass spectrometry.

11.1. Liquid scintillation

Both the samples with high activity of ^{63}Ni and those with low activity were analysed by liquid scintillation. For the first, having an activity of the order of some MBq, the acquisition time was set to 600 seconds, since longer counting times brought the scintillation counter to saturation. For the second type of samples, having an activity of about 20 Bq in 8 ml solution, the acquisition time was set to 3600 seconds, but it could have been longer. For each sample, three acquisitions were performed. The results were analysed in order to find out the variations of activity of ^{63}Ni between the blanks and the corresponding cavitated samples. in particular a negative difference between the cavitated samples and the corresponding blank would indicate a possible reduction of radioactivity due to ultrasonic cavitation and DST reactions. In Table 9 these differences are shown for the high activity samples: Let us remember that the high activity samples were four as reported in chapter 8, Table 3. For the four samples the variations are consistent with each other in the sign, which is always negative, thus indicating a decrease in ^{63}Ni content after ultrasonic treatment. Table 9 reports in the second column a relative difference (in percent), or more explicitly the ratio of the difference mentioned above and the counts per second of the blank:

Table 9: High-activity ^{63}Ni solutions analysed by liquid scintillation. Relative percent variation: difference between the counts per second of the cavitated sample and those of the non-cavitated sample (the blank), divided by the counts per second of the blank, all multiplied by 100.

SAMPLE	LIQUID SCINTILLATION
	relative variation in percent $100 \cdot \left(\frac{\text{cavitated} - \text{blank}}{\text{blank}} \right)$
first sample 13.2 MBq (24th July 2020)	-11.97% ± 0.25%
second sample 2.72 MBq (17th November 2020)	-7.21% ± 0.56%
third sample 2.90 MBq (10th December 2020)	-58.97% ± 0.57%
fourth sample 5.47 MBq (17th December 2020)	-1.05% ± 0.56%

For the low activity samples, on the other hand, no variation of ^{63}Ni could be detected between the cavitated and the blank samples. As mentioned in chapter 7, the beta scintillator that was used (suitable for detecting minute traces of radioactive pollution in the environment and in food), can achieve, through several hours of acquisition, a MDA (minimum detectable activity) of $2 \cdot 10^{-4}$ Bq per millilitre. The low activity samples had 378 Bq of ^{63}Ni in 30 ml and after appropriate preparation, suitable for measurement by liquid scintillation, an activity concentration of $7.56 \cdot 10^{-2}$ Bq per millilitre was achieved, i.e. more than 2 orders of magnitude higher than the MDA.

The reason why no detectable difference was found between the cavitated samples at this very low activity and the blank could either lie in the 3600 seconds, a too short acquisition time to show the variation in activity, or in the too small difference. In fact, it is not yet known if DST reactions have the same transformation efficiency over such a wide range of activity concentration, from the high activity samples to the low activity ones.

11.2. Mass spectrometry

Mass spectrometry is a measurement that does not involve radioactivity. It does not detect ionising radiation and does not produce results in terms of counts of beta particles or X-ray photons per second. ICP-MS counts the atoms discerning among them by their mass. Thus, it does produce results in counts per second, but these counts are the number of detected atoms. Once calibrated by titrated solutions, the counts per second correspond to a concentration. From the equation of the radioactive decay, $A(t)=\lambda\ N(t)$, it becomes clear that the activity of a radionuclide is proportional through the decay probability λ to the number of atoms of this radionuclide. This number, divided by the volume of the solution in which they are dissolved, corresponds to the radionuclide concentration. Therefore, activity concentration and atomic concentration are two proportional quantities. The outcomes obtained by ICP-MS for the high activity samples are reported in Table 10.

Table 10: High activity solutions of ^{63}Ni analysed by mass spectrometry.
Percentage relative change: difference between the counts per second of the cavitated sample and those of the blank, divided by the counts per second of the blank, all multiplied by 100

HIGH ACTIVITY SAMPLE	**MASS SPECTROMETRY (ICP-MS)**
	relative variation in percent $100 \cdot \left(\frac{\text{cavitated} - \text{blank}}{\text{blank}}\right)$
first sample 13.2 MBq (24th July 2020)	-39.1% ± 9.3%
second sample 2.72 MBq (17th November 2020)	-114% ± 28%
third sample 2.90 MBq (10th December 2020)	+19.1% ± 33.7%
fourth sample 5.47 MBq (17th December 2020)	-53.8 % ± 29.6%

The results of the first and the fourth samples are coherent with each other and show a very high decrease of ^{63}Ni in the cavitated sample with respect to the blank. These reductions of ^{63}Ni are also coherent with those emerged by LS. However, it also appears that there is a weird negative value of -114% for the second sample, and a positive variation of ^{63}Ni in the third cavitated sample with respect to the blank. Both the former, above all, and the latter indicate the presence of an experimental problem somewhere along the line of operations and manipulation of the two samples. Before coming up with some hypotheses, it is important to understand how the ICP-MS values in Table 10 are obtained. As it has been said a few lines above, ICP-MS counts the atoms discerning among them by their mass. This means that if there are two different chemical elements with the same atomic mass, they will not be distinguished and will be counted as two atoms of the same isotope. ^{63}Ni has the same mass number of ^{63}Cu which, together with ^{65}Cu, are the two stable isotopes of copper. This element is present in the samples that we cavitated and analysed for two reasons: it is always naturally present at least in trace amounts; it is contained between 1.2% and 2% in the ERGAL, the aluminium alloy of the sonotrode. In the list of isotopes resulting from the ICP-MS analysis, ^{63}Cu is present with its counts per second (cps) and its root standard deviation (RSD). But, from what has been said so far, what is identified by the software as ^{63}Cu is actually the atomic mass 63 (^{63}M) which is the sum of the cps of ^{63}Cu and the cps of ^{63}Ni. The way to mathematically separate the two elements, ^{63}Ni and ^{63}Cu, is based on a reasonable hypothesis and it is as follows. Copper is naturally present on the Earth's crust and it is used in chemistry and/or metallurgy as a chemical element without paying attention to its isotopes. In a certain amount of this natural chemical element its two stable isotopes, ^{63}Cu and ^{65}Cu, show up always with the same percentage abundance: 69.18% of ^{63}Cu and 30.82% of ^{65}Cu. If one knows the quantity of one of them, he/she can always estimate the quantity of the other. In our ICP-MS analyses, ^{65}Cu was the only isotope with the atomic mass 65, thus its counts per second were not mixed up with those of any other isotope. Thus, with the hypothesis that their isotopic abundances be the naturally occurring ones, it becomes possible to know the amount of ^{63}Cu from that of ^{65}Cu. The counts per second of ^{63}Ni are then just the difference between the cps of ^{63}M and those of ^{63}Cu.

$$^{65}Cu \text{ known amount} \quad \rightarrow \quad ^{63}Cu = {}^{65}Cu \frac{69.18\,\%}{30.82\,\%} \quad \rightarrow \quad ^{63}Ni = {}^{63}M - {}^{63}Cu$$

This simple calculation was done to estimate the counts per second of ^{63}Ni in the blank and the cavitated solution for each of the four samples with high activity reported in Table 10. The differences (***^{63}Ni cavitated - ^{63}Ni blank***) relative to the ***^{63}Ni blank*** are the numbers in the right column. Having clarified these simple mathematical steps, let us return to the weird negative variation of -114% of ^{63}Ni. The problem with the second sample arises for the ***^{63}Ni cavitated***. For some reason, which has not been figured out yet, the difference ($^{63}Ni = {}^{63}M - {}^{63}Cu$) is negative. Strangely enough the counts per second of ^{63}Cu (obtained from ^{65}Cu through the isotopic abundances) are larger than those of ^{63}M. It is not easy to put forward hypotheses since the variables to manage are several and their variations may either sum or subtract.

The problems appear in the cavitated sample in which the hypothesis of constant isotopic abundances yields an amount of ^{63}Cu too high with respect to the actual one, which however is unknown. One might imagine that something took place during the cavitation that altered for some reason the relative abundances of ^{63}Cu and ^{65}Cu. Maybe the release of copper from the sonotrode during cavitation was more abundant for ^{65}Cu than for ^{63}Cu, or maybe one could imagine that the DST reactions might have affected the two copper isotopes in a different way so to generate an unnatural imbalance between them. In any case, these two anomalies (second and third samples) pushed the experimenter to reconsider all the operations that had been done in preparing the samples and in treating them by ultrasound in order to find out in the experimental notes some possible mistake in the steps of the protocols. So far, no inconsistencies were found in the reproducibility of the experimental conditions. However another consideration has to be presented. The last steps of the experimental protocols always involved the preparation of the blank and cavitated samples for ICP-MS and LS. While the samples prepared for LS were not manipulated any further after being sealed and were inserted in the apparatus of LS analysis exactly as they were received, unfortunately this was not the case for ICP-MS analyses. In particular if the concentration of the atoms to detect in the samples is high (ICP-MS is sensitive down to 1 ppb and lower), the samples get diluted. In other words, this means that they are not inserted in the apparatus of analysis exactly as they were produced, but that further manipulations, which are not part of the controllable protocols, are carried out on them. This consideration does not imply anything about the quality of the ICP-MS analyses, it means only that in trying to pinpoint the problem in the whole history of the two "anomalous" samples these manipulations are the only part that slips away from our control.

Of course, the mass spectrometry was also used to analyse the samples of stable atoms, Ni, Cu and Fe. Of all the samples listed in Table 7 and Table 8 in chapter 11, some samples haven't been analysed yet, in particular, some samples of copper and all those of iron, so the results are not complete. The results of the ICP-MS analyses are reported in the following Table 11:

Table 11: Results of the ICP-MS for the stable nuclei Ni and Cu.

Sample of stable nuclide	**Mass spectrometry**
	relative variation in percent $100 \cdot \left(\frac{\text{cavitated} - \text{blank}}{\text{blank}}\right)$
^{60}Ni solution 1 ppm	-15.98 % ± 1.65%
^{60}Ni solution 5 ppm	-14.36 % ± 1.64%
^{60}Ni solution 10 ppm	-3.35 % ± 0.73%
^{60}Ni solution 1000 ppm	+31.70 % ± 2.56%
-	-
^{63}Cu e ^{65}Cu solution 5 ppm	not meaningful
^{63}Cu e ^{65}Cu solution 10 ppm	not meaningful
^{63}Cu e ^{65}Cu solution da 1000 ppm	-10.20 % ± 2.81% e -10.05 % ± 2.67%

The isotope 60 of nickel is the second most abundant of its stable isotopes. The variation of the number of atoms in the solutions after the application of ultrasound is rather important and for the first three initial concentrations, 1, 5 and 10 ppm, they are negative, that is the ultrasound caused a decrease of the atoms of ^{60}Ni. On the other hand, the variation of ^{60}Ni in the 1000 ppm solution is even larger but positive. This should not surprise, despite the lack of complete coherence among the samples of different concentrations. The main result to be considered is the variation of concentration of ^{60}Ni by ultrasound. These mechanical compressions and decompressions should be accounted for a variation, either positive or negative, of the number of atoms of an atomic species. As to the negative variations, it is of course necessary to verify whether the disappearance of the atoms from the solution is not due to their adsorption on the walls of the glass and the sonotrode, which thanks to the very low pH is rather unlikely. As to the positive variation, this is even larger and more astonishing since nickel is neither part of the composition of the glass, nor part of the sonotrode alloy, nor it was present in the experimental equipment.

Further more, the inconsistency in the sign of the variations between 1, 5, 10 ppm and 1000 ppm is only due to the lack of knowledge of the DST reactions whose behaviour is still almost completely unknown.

With regards to the variations for the copper, there are no positive results for the 5 and 10 ppm. In other words, the variations of the number of atoms of copper are within the uncertainty of the initial concentration. Moreover one has to consider that copper might also come from the sonotrode that contains copper between 1.2 and 2 %. In this sense, all the positive variations that were obtained were, for this reason, disregarded. On the contrary, for the 1000 ppm solution, the variation is negative which is a remarkable thing because, despite the possible release of copper from the sonotrode the number of its atoms decreased after cavitation.

11.3. Bremsstrahlung X-ray spectrometry

X-ray spectrometry of the Bremsstrahlung of the ^{63}Ni beta particles in the solution and in the glass of the vessel is the most indirect technique to measure ^{63}Ni content. This radiation appeared to be centred around 30 keV with a very broad peak as in Fig.4.

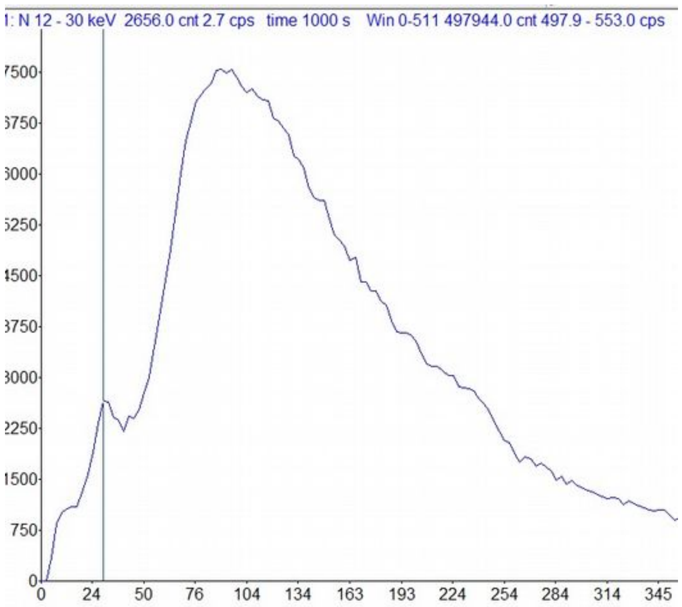

Fig.4 Spectrum of ^{63}Ni blank by the Atomtex 6102A NaI spectrometer. The Bremsstrahlung emission is the small broad peak on the left where the marker is. The peak is centred at 30 keV.

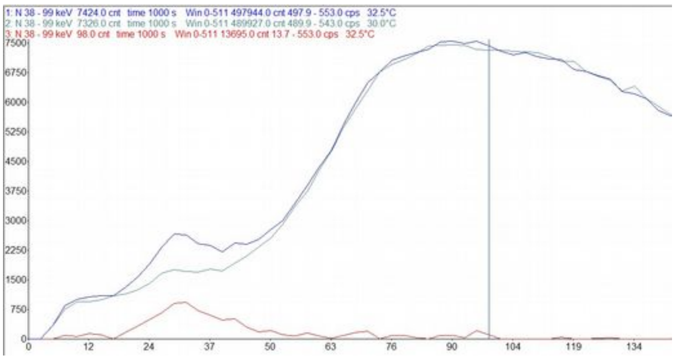

Fig.5. Spectrum of ^{63}Ni blank in blue, background in green and their subtraction in red by the Atomtex 6102A NaI spectrometer

In Fig.5 both the ^{63}Ni blank and the background and their subtraction are shown. One might argue that the background is rather high. This is due to the decision to fix once for all the position of the cavitation vessel in the ultrasound machine so that both before and after cavitation the detection geometrical efficiency would remain constant. However, this decision implied the impossibility to use the lead shielding of the detectors since the ultrasound machine would have not fitted in it. Thus it resulted in a rather high X-ray background, which did not allow for low measurement uncertainties in the 1000-second time intervals in which the spectra were acquired. The comparison by difference of the areas of the spectrum zones affected by Bremsstrahlung between the cavitated and blank reference sample always showed a systematic decrease of the radioactivity of ^{63}Ni after ultrasonic treatment but unfortunately, all of them were affected by very high uncertainty. For this reason, in Table 12, only two differences between cavitated samples and the corresponding blank are significant. The correspond to two of the solutions with high activity of ^{63}Ni. As to the low activity samples, no X-ray measurements were performed since the Bremsstrahlung is so low that not even long acquisition intervals would show anything different from the X-ray background.

Table 12: Highly active ^{63}Ni solutions analysed by X spectrometry.
Relative percent change: difference between the counts per second of the cavitated sample and those of the blank, divided by the counts per second of the blank, all multiplied by 100

Sample	**BREMSSTRAHLUNG** **X-rays**
	relative variation in percent $100 \cdot \left(\frac{\text{cavitated} - \text{blank}}{\text{blank}}\right)$
first sample 13.2 MBq (24th July 2020)	-4.7 % ± 1.3%
second sample 2.72 MBq (17th November 2020)	-10.7 % ± 10.3%
third sample 2.90 MBq (10th December 2020)	negative but not significant
fourth sample 5.47 MBq (17th December 2020)	negative but not significant

Chapter 12.

Study and evaluation of the possible spurious factors that may produce a variation of the radioactivity and/or the concentration of the radioactive or stable nuclei

The results that have been shown in the previous chapter are all variations of the activity of ^{63}Ni or variations of its concentration or variations of concentration of stable nuclides. However other processes other than ultrasound and space-time deformation reactions may also cause changes of the concentration of ^{63}Ni and of other atoms. They can introduce variations that are not those that have been looked for. Four main processes have been identified and somehow analysed in order to estimate the percentage of variations either positive or negative that they may bring about and then apply the necessary corrections.

12.1. Natural radioactive decay of ^{63}Ni

The first process considered was the spontaneous natural decay of ^{63}Ni into the stable nuclide ^{63}Cu. The half-life of ^{63}Ni, i.e. the time interval in which starting from a certain amount of this radionuclide one arrives at half the amount, is about 100 years. This is precisely one of the reasons why this radionuclide was chosen, as this long half-life would have allowed us to neglect its decrease during all the measurement processes, which always lasted a maximum of ten hours per experimental day. In doing a quick calculation to figure out how much it is the percentage of decrease of ^{63}Ni in 10 hours due to natural radioactive decay, one obviously uses the exponential law of decay with the decay constant relative to ^{63}Ni. In doing this, the percentage of decrease of ^{63}Ni has an absolute value of 0.0008%, which is completely negligible compared to the measured percentages of change shown in the tables in the previous chapter.

12.2. Sampling of the radioactive solutions between two consecutive cavitations to prepare the samples for LS

The second cause of possible variation of ^{63}Ni between before and after ultrasonic treatment (a cause that has actually been shown not to exist) are the samplings of cavitated solution between cavitations (ultrasonic treatment). These microlitre amounts of solution were taken to prepare the samples to analyse by liquid scintillation and thus to monitor the change in ^{63}Ni due to ultrasound and the DST reactions between ultrasonic treatments. By taking these small amounts of solution, it seemed at first glance that by subtracting a certain number of ^{63}Ni atoms per sample, these subtracted atoms should be taken into account when comparing the ^{63}Ni concentrations between the various cavitated and blank (non-cavitated) samples. However, these samples only varied the total number of ^{63}Ni atoms in the samples, but had no effect on the concentration of ^{63}Ni, which remained the same. In fact, the samples were taken from homogeneous solutions (as per the initial chemical preparation and ultrasonic mixing), whose concentration always remained the same and obviously in comparing the different cavitated and blank samples, the volumes of solution analysed by ICP-MS and LS were always the same.

12.3. Water evaporation due to the increase of temperature during cavitation

The third process that could lead to a change in ^{63}Ni concentration other than nuclear DST reactions is the evaporation of the solution during ultrasonic treatments due to an increase of the temperature. In order to estimate these percentage changes in ^{63}Ni concentration due to evaporation, it is very reasonable to consider that the evaporation process involves only water, i.e. only the water molecules evaporate without involving ^{63}Ni or the other atoms. This reasoning stems from the fact that, first of all, the solutions are at very low pH, as mentioned above, and that this implies that the atoms of ^{63}Ni are present in solution in ionic form as ^{63}Ni+ and therefore more strongly interacting with the $(NO_3)^-$ anions of the nitric acid than the water molecules linked to each other by the weaker hydrogen bond. Furthermore, none of the substances present in solution are organic substances, which would have greater volatility. Based on this very reasonable hypothesis, but having had so far no experimental opportunity to verify it, the changes in concentrations of ^{63}Ni due to evaporation were calculated. Gravimetric measurements of the solution between the various cavitation processes were always present in the measurement protocol, precisely in view of these evaporation estimates to be performed.

Without presenting the change in concentration of ^{63}Ni for each solution, we report the extremes obtained. The percentage change in ^{63}Ni concentration due to evaporation of water from the solutions is between 0.07% and 0.17%. It should be noted that these values are lower than the percentage changes obtained with LS and ICP-MS. These variations of ^{63}Ni experimentally obtained do contain the variation due to the evaporation which must therefore be reintegrated. In this connection it should be noted that the evaporation of water increases the concentration of ^{63}Ni (i.e. 0.07% and 0.17% are concentration increases). Therefore, since the experimental data show a decrease of ^{63}Ni, it means that this decrease is actually greater, i.e. that these estimates of the evaporation must be added to the experimental values, thus leading to a larger percentage decrease of ^{63}Ni.

12.4. Absorption of the radioactive and/or stable nuclei on the glass walls of the vessel and/or on the surface of the sonotrode

A further possibility of variation in the amount of ^{63}Ni is that it may be adsorbed by the walls of the sonotrode and those of the glass vessel. Although it is very unlikely that such an adsorption could take place due to the negative pH values of the cavitated solutions, which prevent this possibility from happening, it would still be appropriate to carry out surface analyses, e.g. by means of X-ray microanalysis of both the sonotrode and the glass container. This experimental check was planned but unfortunately not performed yet due to a lack of time.

Chapter 13.

Concluding Remarks
Comparison of the data obtained by the three different measurement techniques

In Table 13, the relative percentage changes of ^{63}Ni obtained by the three measurement techniques used are shown together for comparison: X-spectrometry, mass spectrometry and liquid scintillation.

Table 13: Comparison of variations between the three analysis techniques

	percentage relative variation of the activity of ^{63}Ni		
sample	**Bremsstrahlung X-rays**	**ICP-MS**	**LS**
first sample 13.2 MBq (24th July 2020)	**- 4.7 % ± 1.3 %**	**-39.1% ± 9.3%**	**-11.97% ± 0.25%**
second sample 2.72 MBq (17th November 2020)	**-10.7 % ± 10.3 %**	**-114% ± 28%**	**-7.21% ± 0.56%**
third sample 2.90 MBq (10th December 2020)	**negative but not meaningful**	**+19.1% ± 33.7%**	**-58.97% ± 0.57%**
fourth sample 5.47 MBq (17th December 2020)	**negative but not meaningful**	**-53.8 % ± 29.6%**	**-1.05% ± 0.56%**

Apart from the anomalous numbers highlighted in red in the two central cells of the ICP-MS column, -114% and +19.1%, the data from the three measurement techniques, based on completely different measurement methods, agree with each other in showing a decrease of ^{63}Ni after ultrasonic treatment. The problem that exists in the ICP-MS column for the two anomalous numbers will be thoroughly investigated. The negative variation, -114%, of ^{63}Ni is almost certainly a mistake and it is more likely due to the manipulations before ICP-MS analysis, rather than in the preparation of the samples. As to the positive variation, +19%, it also appears a mistake in the ICP-MS analysis. In fact, all the various experimental procedures, preparation of the samples, application of ultrasound, sealing of the samples to be analysed, which were meticulously followed and painstakingly reported in the laboratory notes, have been controlled several times without showing any indication pointing out any anomaly in them. It also seems unlikely that these two anomalies may be intrinsic to the studied phenomenon, the DST reactions. But anyway not only the ICP-MS analyses have been reprogrammed to be repeated soon, but furthers test on radioactive solutions with the same characteristics of the second and the third samples will also be repeated.

Chapter 14.

Conclusions

In order to make more tangible the reduction of activity (concentration) of ^{63}Ni reported in Table 13 and hence to make clearer the possible huge advantage of this technique in treating radioactive wastes, let us consider a decrease of ^{63}Ni of 40%, a hypothetical variation close to the highest variations registered by ICP-MS or LS. In Fig.6 two exponential curves are shown. The one above (in blue) belongs to the ^{63}Ni which naturally decays and whose quantity decreases of 40% in almost 80 years. The curve below, instead, (in red) represents the decrease of ^{63}Ni which is treated by ultrasound at the beginning for 600 seconds. Of course this short interval of time cannot be appreciated in such a scale of hundreds of years, thus the decrease seems vertical and taking place in no time. This is not the case, but 600 seconds is an interval $2 \cdot 10^{-7}$ times shorter than 80 years.

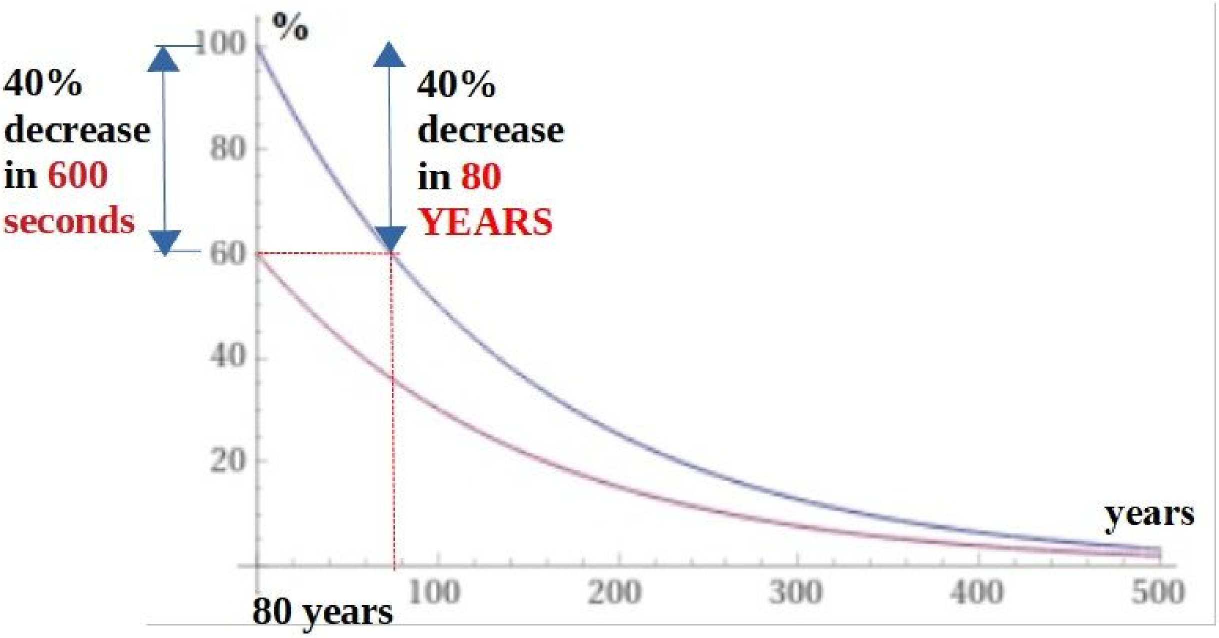

Fig.6. This figure shows the radioactive natural decay of ^{63}Ni with the exponential decay curve above (in blue). In 80 years the decrease is about 40 %. In the exponential curve below, in red, it is shown the same amount of ^{63}Ni which is treated at the beginning by ultrasonic cavitation and in 600 seconds (not visible on the scale of hundreds of years) decreases of 40% as well.

As it has been shown by several experiments since almost 3 decades so far [32,33,39,40,41,42] there exists the possibility to treat radioactive materials by not so complicated equipments, all involving pressure, in order to transform them into neutral ones. These experiments are not at the level of industrial prototypes but all of them indicate with concordant results that this deactivation is indeed possible. Many criticisms are still raised, as to the reproducibility of the results and the impossibility of these results to occur because they are impossible according to the present theoretical knowledge of physics. But this should not discourage to carry on the research. When Rutherford discovered the nucleus of the atom he himself had no doubt that the nucleus was so hard that it could and would never be destroyed. But thank goodness knowledge does not stop and these phenomena that appear anomalous and impossible now may be our future technology as it is nowadays the "impossible" nuclear fission. The only hope that dwells in the minds of those people who are involved in researching and understanding these new phenomena is that with the time an always growing number of researchers will be involved in this fascinating research that leads not only to a higher knowledge and contemplation of Nature but also leads to new technologies that become more and more important each day for our survival and that of the new generations.

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