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DST-deactivation of nickel-63 nitrate

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Abstract: Recent theoretical and experimental results, based on an extension of the Einstein theory of relativity, show that nuclear reactions of a new type can occur. In this framework, the feasibility of the deactivation of radionuclides is investigated. This paper reports the deactivation of nickel nitrate made of radioactive Nickel-63 in nitric acid by using ultrasounds. From the applicative point of view, it is a more realistic system then the previously considered thorium, not only because the molecule and the system are more complex but also because the problems related to the high corrosion and radioactivity have been treated at the same time, thus miming realistic situations like those of deactivation of nuclear waste.

Keywords: Deactivation of radionuclides, ultrasounds, deformed space time, Local Lorentz Invariance breakdown, Nickel-63.

While we were writing the present work,
Professor Roberto Mignani,
"THE" Theorist of the Deformed Space-Time theory,
passed away.
We dedicate to him this article,
one of the many
based on the Deformed Space-Time theory.
The authors

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1 Introduction

Transformations of matter induced by ultrasound (US) irradiation are reported in water [1–3] while neutron emissions have been measured from water solutions [4, 5] and solid material [6, 7]. By following the Deformed Space Time (DST) theory [8, 9], they have been explained as consequences of nuclear reactions produced by an appropriate concentration of energy in space and time [10]. Moreover, evidences of nuclear transformations by DST-reactions were also obtained in mercury [11–13].

These evidences just mentioned show the existence of a new type of nuclear reactions that have nothing to do with any of the well-known nuclear processes. Hence on the base of these experimental evidences one may assume that these reactions can also affect radioactive nuclides transforming them into non-radioactive ones without following the normal radioactive decay and without using processes involving apparatuses of large dimensions. In fact, a radioactivity decrease larger than that expected by considering its emission rate was reported for thorium-228 [14].

Reduction of the activity is the main goal in the treatment of waste from nuclear plants. It is also important in the treatment of materials from hospitals and for the purification of drinking water.

Concerning the drinking water, its radioactivity is strictly related to that of the earth which is about 85 % natural and 15 % artificial [15]. Different techniques for different contaminants are used in this treatment. The most used ones [15, 16] are ion exchange, reverse osmosis [17], distillation, aeration and adsorption by granular activated carbon or, more recently, oxidatively modified carbon [18]; good results were obtained by using graphene oxide [19].

Similar treatments [20] are also used for contaminated water of nuclear plants.

The vast problem of decontamination of nuclear plants is mainly faced by using two families of methods: mechanical methods and chemical/electrochemical methods. The first ones are mainly used for single components while the second ones find a wider field of application.

However, all the mentioned methods do not modify the activity of the single nucleus. They only separate the active part from the remaining material.

The same "deactivation" of the active part usually consists in incorporating it in a larger volume of inert material [21], thus decreasing its activity per unit volume, but the total number of radionuclides is not modified.

A real deactivation, able to reduce the total number of original radionuclides in a shorter time with respect to the natural decay, is not yet considered on a large scale.

However, some efforts are reported in literature. On the theoretical side, for instance, the effect of electron environment on the probability of barrier penetration and on the alpha decay was studied [22] by replacing the nuclear potential with a Fermi-Thomas potential for heavy atoms while a potential derived from a constant electron-density model was used for very light atoms.

In a subsequent study the increase of the electron density was predicted [23] to shorten the decay time of alpha decay.

Experimentally, the effect of electrons was shown [24] to depend on the entrance channel of the nuclear reactions and not to be influenced by the emitted particles, so suggesting a possible new way to enhance the alpha decay rate of transuranic waste.

However, it is evident that problems of handling and radioprotection are not eliminated after this enhancement.

The effect of the environment was also studied [25] in the alpha decay of Fr-221 at room temperature in different materials: gold, tungsten, silicon and polyethylene. Changes – smaller than 5 ‰ – were found depending on the solid-state environment.

Beside the environment, the temperature effects were investigated: no appreciable difference was observed either in the half-life of beta decay of Au-198 at room temperature with respect to 12 °K [26] or in the alpha decay of Po-210 implanted in a copper matrix at 4.2° or at 293° K [27].

These studies have all been searching for new possibilities to reduce the radioactivity and hence for phenomena and conditions that are new with respect to the consolidated physical knowledge. Nevertheless, the ultimate theoretical background common to each of them is based on the Minkowski space-time, i. e. a flat and isotropic space-time.

In the case of the DST nuclear reactions, on the contrary, a non-minkowskian space-time, i. e. a deformed space-time, is present and plays the key role in them. In this sense, a non-traditional path of decay of radioactive nuclides is expected to exist [28] and its signature is looked for.

Beside the evidence already obtained with Th-228 [14], further observations are required to study this unusual deactivation, we named DST-deactivation to stress that it is a consequence of a space-time deformation, as predicted by the DST-theory.

To clarify this new concept, an experiment was conceived and realized. In the following pages, we will not deal with this semantic problem any further but rather we will focus our attention on the effects induced by DST-reactions on a radioactive material when these reactions are obtained by using ultrasound and cavitation.

For the sake of completeness, we recall that if DST-reactions occur then the Lorentz Local Invariance (LLI) is violated. More details on this subject can be found in literature [8, 9, 29].

The studied sample is nickel nitrate ($Ni(NO_3)_2$) made of radioactive Nickel-63, in nitric acid (HNO_3). This sample is more complex and more realistic than the pure thorium solution studied in the past [14]. As the radionuclide is a part of a molecule also containing other elements. This fact makes the conclusions of more general application. In addition, the sample is very corrosive, as it is the case of most nuclear waste, either of industrial or sanitary source, that are the most critical to be treated. Thus, the reported experiment can be considered as a positive test for future applications in severe conditions: radioactive and corrosive samples.

2 The experiment

Radioactive Nickel-63 in nickel nitrate molecules dispersed in nitric acid (67 %, 1.5 molar) were sonicated by using 35 KHz ultrasounds (US-irradiation or sonication). A scheme of the experimental set-up is reported in Figure 1.

Nickel-63 is a beta-emitter, 66 keV (10^{-2} fJ) being the maximum energy of the beta electron.

The sample total mass was 39.6 g, the total volume was 30 cm^3: 25 cm^3 of nickel nitrate molecules in nitric acid and 5 cm^3 of bi-distilled and deionized water.

The sample was sonicated in two runs of 100 s each. The amplitude of the sonotrode tip oscillations was 7 μm. The sonotrode was the Aluminum alloy ERGAL 7075.

The geometry of the sonotrode and sample vessel for sonication was chosen in accordance with the parameters imposed by Lorentz violation in the framework of the DST theory, as done in former DST-reactions: for instance, when nuclear reactions of new type occurred in mercury [11–13].

Vessels of different shape and sonotrodes of different material were used. The increase of temperature was assumed as an index to evaluate the efficiency of the

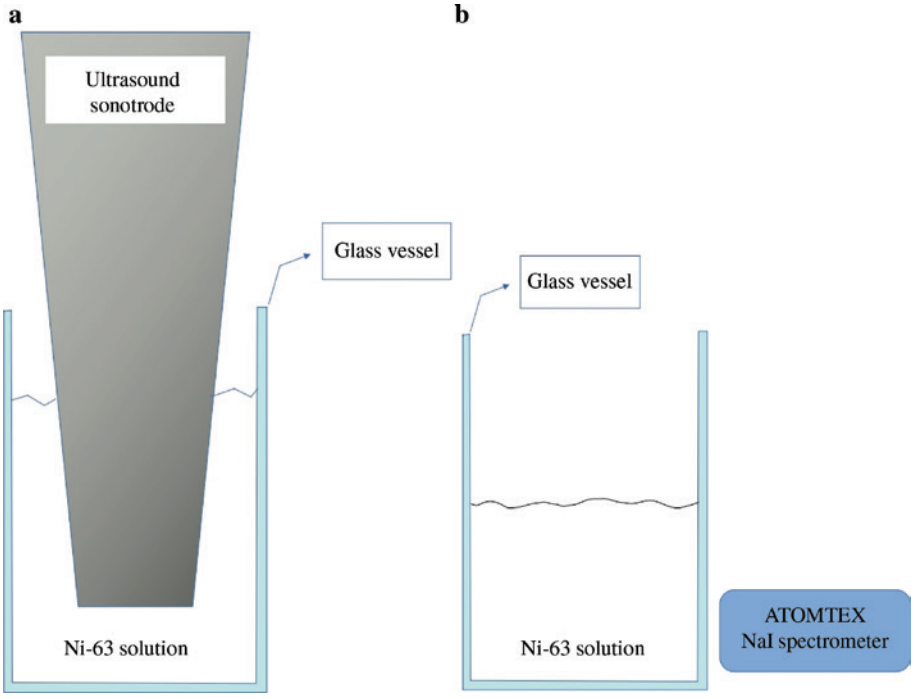

Figure 1: Sketch of the experimental set-up.
(a) During the sonication. (b) During the X-ray measurement. (A more detailed scheme cannot be provided because of a pending patent.)

whole apparatus vessel/sonotrode. When the highest temperature increase was obtained, the below reported measurements took place.

A dosimeter-ratemeter (S.E. International, Mod. Radiation Alert), a Geiger counter (model Gamma Scout™) and a portable NaI spectrometer ATOMTEX (Model AT6102, n.61154) were used to detect the emitted radiation. They measured the total intensity or counts of beta, gamma and X-rays. The NaI spectrometer, placed at 0.5 cm apart from the sample, recorded the energy spectrum of the X-rays emitted by Bremsstrahlung by the beta particles emitted by Ni-63.

Although an exhaustive description of the experimental set-up cannot be provided in this paper for patent reasons, it is useful for the reader to know that the actual measurement, aimed at detecting the reduced activity of the solution, was carried out by the NaI X-ray and gamma spectrometer. As a matter of fact, it was impossible to detect directly the beta electrons emitted by Ni-63, because even the most energetic ones at 66 keV were stopped by 1 cm of water, the thickness of the vessel, and the case of the NaI spectrometer. It is easily verifiable, for example through the application ESTAR of the NIST that electrons of 66 keV are completely stopped by 0.18 cm of glass and a bit more than 0.3 cm of water and of polyethylene which may simulate just the external case of the NaI spectrometer. In this sense, the spectra shown in the following figures can be only interpreted as the Bremsstrahlung X-rays of the beta electron of Ni-63.

The dosimeter monitored only the dose and dose-rate in the laboratory. The Geiger counter, on the other hand, having the possibility to measure without any screen but the thin beryllium window, was used to monitor the possible contamination of the surface surrounding the vessel and of the surface of the sonotrode. No significant variation with respect to the background in any part of the experiment was registered by any of these two instruments. For this reason, their measurements will not be reported in the following.

The whole measurement of the radiation emitted by the sample can be divided into three parts:

- Measurement of the gamma/X radiation spectrum emitted by the sample before sonication;
- Measurement of the gamma/X radiation spectrum emitted by the sample after 100 s of sonication at a frequency of 35 KHz and with a transferred power of 15 W, corresponding to a vibration amplitude of 7 microns;
- Measurement of the gamma/X radiation spectrum emitted by the sample after an additional sonication run of 100 s.

3 Results

In the first part of the experiment, the X/gamma background spectrum of the laboratory was measured by the NaI spectrometer before bringing the radioactive sample into it.

The second step was the measurement of the radiation emitted by the sample before sonication during a time interval of 100 s again by the NaI spectrometer.

After 100 s of sonication, the radiation measurement of the sample was repeated and so it was again after an additional sonication run, always of 100 s. Finally, a further measurement of the X/gamma background spectrum was performed again.

The background spectrum before the sample measurements (Figure 2) and after are comparable: for each channel of the spectrometer the difference between the counts before and after is well lower than the statistical error.

The Figure 3 reports the full spectrum before sonication. It contains both the background and the Bremsstrahlung peak of the electrons emitted from Ni-63. This peak is centered at 30 keV.

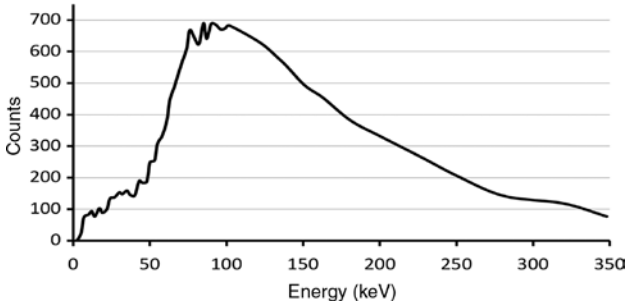

Figure 2: Spectrum of the background radiation measured by the NaI spectrometer

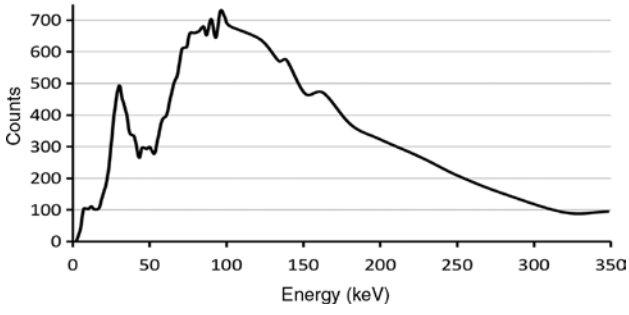

Figure 3: Background spectrum with the superimposed peak between 0 and 50 keV of the Bremsstrahlung radiation due to the electrons from the beta decay of Ni-63 measured by the NaI spectrometer before sonication.

The Figure 4 reports the same spectrum after 100 s sonication.

By the way, we like to note that Ni-63 has been chosen because its Bremsstrahlung peak occurs in a spectral region where the background is low and nearly linear.

After subtracting the background, the intensity spectra of the X-rays emitted by Bremsstrahlung by the electrons from beta decay of Ni-63 were obtained and are reported in Figure 5. The energy axis ranges from 0 to 66 keV, which is the maximum energy of the beta-electrons from Ni-63.

The Figure 5a reports the bremsstrahlung spectrum before ultrasound-irradiation; the Figure 5b reports the spectrum after 100 s of sonication and Figure 5c after additional 100 s of sonication.

It is useful to spend a few words on what these figures show. They are the detected X-ray Bremsstrahlung spectra obtained from the beta electrons that, after their emission by the Ni-63 nuclei, radiate as they lose their energy in moving through the acid solution containing the Ni-63 and through the glass walls of the vessel.

The beta energy spectrum emitted by Ni-63 can be obtained by the theory of beta decay of nuclei and statistical considerations about the phase space of the electron and neutrino energy. It ranges from a few keV up to 66.7 keV with a maximum probability of emission at about 17 keV.

The conservation of energy in the Bremsstrahlung process dictates that the X-ray spectrum must have the same features if the X-rays are not attenuated in passing through some layers of different materials. This is not the case in the present experimental set-up as the X-rays are subjected to attenuation.

Thus, the shape of the X-ray spectrum recorded by the NaI spectrometer depends on the energy of the

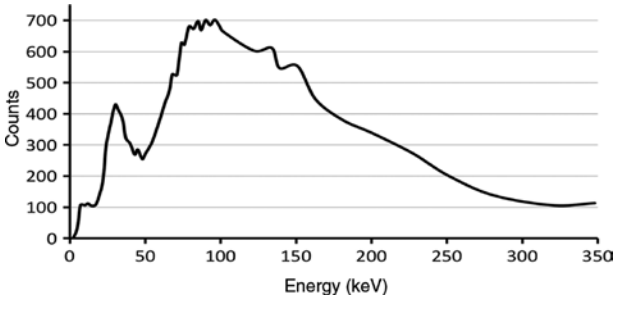

Figure 4: Background spectrum with the superimposed peak between 0 and 50 keV of the Bremsstrahlung radiation due to the electrons from the beta decay of Ni-63 measured after 100 s of sonication.
(NaI spectrometer).

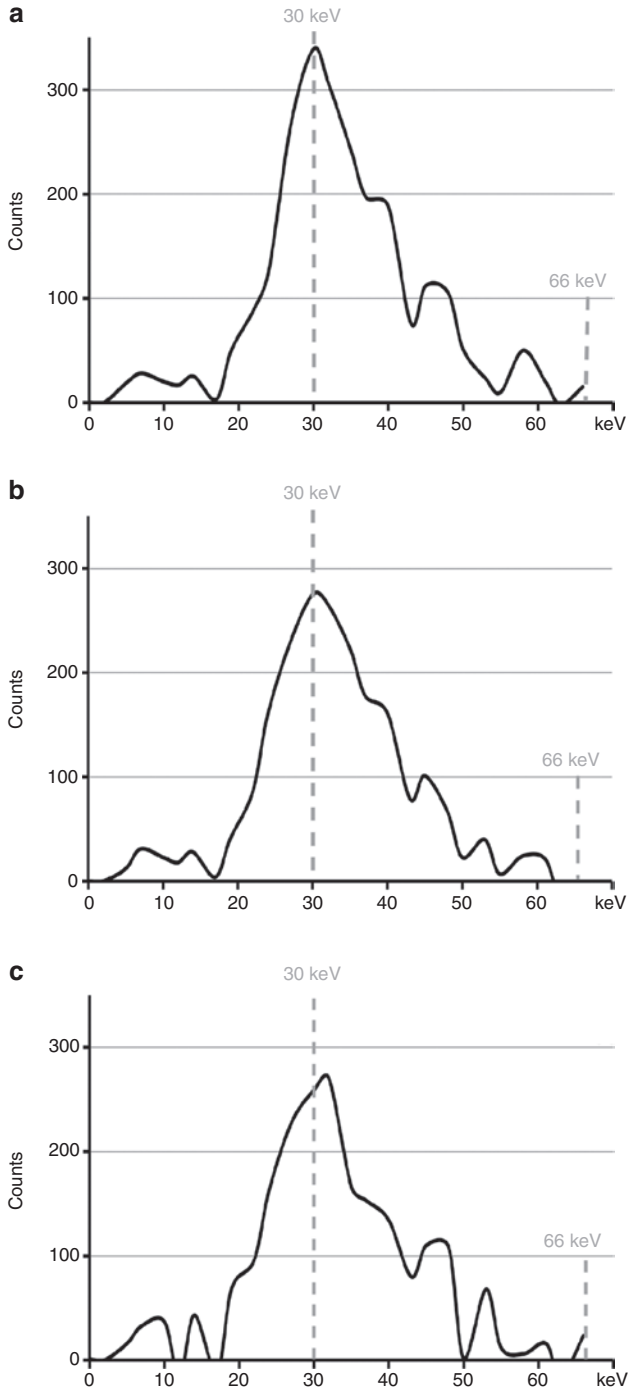

Figure 5: The spectra of bremsstrahlung radiation produced by beta electrons emitted by Ni-63.
(NaI spectrometer). (a) before sonication; (b) after 100 s of sonication (35 KHz, 15 W); (c) after additional 100 s of sonication (35 KHz, 15 W).

Bremsstrahlung photon; the number of photons for each energy; the composition and the thickness of the material they go through; and the intrinsic detection efficiency of the spectrometer.

The absorption can be evaluated, for instance by using an online X-ray transmission calculator, by only considering the thickness of the vessel (about 2 mm). Although the X-ray photons produced by Bremsstrahlung at 17 keV are more abundant than those at 30 keV, the 17 keV photons are more attenuated than those at 30 keV. If one also considers the contribution of water, which is order of magnitude lower than that of the glass, and of hydrochloric acid, present in solution, besides the strong contribution due to the efficiency of the NaI spectrometer, characterized by an abrupt decrease at energy lower than 20 keV, the effect is still more enhanced.

These facts make sense of the shape of Bremsstrahlung peaks in Figure 5, where the intensity at 17 keV, corresponding to the maximum of the emitted photons, is much lower than at 30 keV, maximum of the detected photons.

To point out the changes induced by the US-irradiation, the total area of each peak was evaluated from 0 to 66 keV: the results are reported in Table 1.

The error was evaluated from the statistical fluctuations of the bremsstrahlung spectrum with background and of the background without bremsstrahlung peak in the range from 0 to 66 keV.

4 Discussion

The most amazing result is the sum of counts from 0 to 66 keV, which decreases of about 13 % after 100 s of ultrasound irradiation: it is reported in the first and second rows of Table 1, which concerns the integrated intensities of Figure 5.

A time-lapse of 20 years would be necessary to obtain a similar decrease according to the lifetime of the Ni-63 nuclide, having a half-life of 101.2 ± 1.5 years [30].

This result is a more accurate confirmation of the results obtained in the case of thorium [14]: also in that case a decrease of radioactivity was observed after ultrasound irradiation but the experimental incertitude on the obtained ratio was larger than in our case.

Table 1: Total area of the bremsstrahlung peak from 0 to 66 keV before sonication, after 100 s of sonication and after 100 + 100 s of sonication.

Peak integral before sonication	2380 ± 110
Peak integral after 100 s of sonication	2070 ± 110
Peak integral after 100 + 100 s of sonication	2046 ± 110

Although more accurate, the present test aims to detect the occurrence of the phenomenon, while more precise results, obtainable over longer times, are left to future measurements, where the improvement of the statistic aspect is the main goal.

It is of interest to note that the additional irradiation of 100 s resulted in a further decrease of the counts but of lower entity, as it can be deduced from the second and third row of Table 2. In fact, the total decrease after the two pulses is about 14 % of the initial value.

To take into account this latter behavior, we can suppose that some competitive effects take place. To discuss this point, we remark that the experiment was conceived on the footsteps of previous experiments [11–13, 31–34] in which the space-time deformation was supposed to occur. This deformation can only be produced by an appropriate concentration of energy in space and time [9, 29, 34]. Besides, the nuclear reactions were observed to occur inside micro-reactors, such as micro-cavities in solids (so-called "Ridolfi cavities" [35]) or micro-bubbles in liquids [28].

In our case, beside the appropriate geometry of the experiment, the suitable distilled water was added to obtain the right population of useful microbubbles working as micro-reactors by sonication. By the way, this is a part of the technological procedure.

In this framework, the cause of the lower decrease of radioactivity after the further 100 s of sonication can either be other primary reactions with longer latent time or secondary reactions following the primary ones that absorb some of the energy necessary to deform space-time or also the disruption of the micro reactors, with consequent reduction of the bubble population available for the next reaction runs.

However, the experimental error on these percentages (±6 %) makes pointless any further comparison between the two runs, although the difference between sonicated and not-sonicated sample is confirmed.

If a deformation of space-time is assumed, then we could also consider the decreased intensity as a consequence of a time deformation of the weak interaction – responsible for the nuclear decay – with respect to the electromagnetic interaction, which is fundamental in all the electronic devices and in our perception of the external world. However, up to the present date, no relative deformation of time between the two interactions is reported in literature, although time deformation is described in the case of other interactions [8, 9].

As a support to the observed phenomenon of the anomalous behavior in the decay of a radionuclide, we consider, beside the already cited case of thorium [14], also the observations collected by P. Hagelstein and collegues at the Massachusetts Institute of Technology, Cambridge, MA, USA [36]: they recorded an anomalous decay in X-ray and gamma emission of a radioactive isotope of Cobalt in an experiment with ultrasound irradiation.

4.1 Consequences on the decay

The dashed line in Figure 6 corresponds to the expected count rate decrease as a function of time by considering the decay constant of Ni-63.

In the same figure, the continuous line is characterized by a down step at short times which corresponds to the decrease of counts registered after sonication. We may assume that this down step is the consequence of the reduced number of radioactive nuclei transformed by the DST reactions into inert nuclides. Since we also assume that the decay constant is not affected by sonication, the continuous curve approaches asymptotically the dashed one at long times. However, if we consider the time corresponding to half of the initial number of Ni-63 atoms, the value obtained after sonication is more than 20 years shorter than the natural decay without sonication. Thus, we can state that in less than 5 min the half-life was

Table 2: Experimental details in thorium experiment and in Nickel experiment.

	Thorium experiment (Ref. [14])	Nickel experiment
Ultrasound frequency	20 KHz	35 KHz
US transferred power	130 W	15 W
Amplitude of the sonotrode tip	30 microns	7 microns
Sample mass	300 g	39.6 g
Sample volume	300 cm^3	30 cm^3
Sonication runs	1 run of 90 min	2 runs × 100 s
Sonotrode composition	Steel alloy	Aluminum alloy
Sample container	Reaction chamber	Vessel

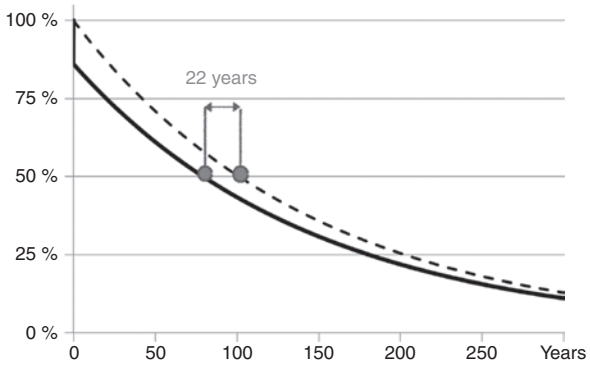

Figure 6: Decay of Ni-63.
Dashed line: natural decay. Continuous line: after 100 + 100 s of sonication (35 kHz, 15 W).

shortened by about 20 years … although the decay constant is the same!

The transformation by sonication of radionuclides into stable nuclides in a shorter time than the usual radioactive half-life is an effect that could occur also in the above-quoted anomalous decay of Cobalt [36]. Other effects can also occur in the cobalt experiment: for instance, radionuclides with different decay time can be transformed with different latent time after the sonication. In any case, the time behavior of emitted energy appears to be anomalous due to DST-reactions.

Thus, the quoted Cobalt experiment and the present Nickel experiment confirm each other.

4.2 Comparison with thorium experiment

About 10 years have passed from the thorium experiment [14]. What has been learnt in the meantime?

To answer this question, let's consider first the differences between the two experiments, as reported in Table 2.

The geometry of both the sonotrodes was chosen by following the rules considered mandatory for inducing the violation of Local Lorentz Invariance and hence the Space-Time deformation.

The sample container, on the contrary, was chosen without following this kind of rules in the case of thorium, while, for Nickel, it is a vessel with an appropriate geometry. These geometric rules aim to obtain an appropriate distribution of energy concentration. In fact, the nuclear DST-reactions were observed to occur or not to occur depending on the energy distribution in the previous experiments with Mercury [11–13].

Both for Thorium and for Nickel, the micro-reactors are the bubbles of appropriate size contained in the solution and made collapse by sonication and cavitation.

In conclusion, the main difference between the two experiments consists in the geometry of the container. i. e. the vessel rather than the reaction chamber. In terms of acquired know-how, this means that the reactions in thorium were produced just by choosing the appropriate bubble size, while the chamber geometry design converged on the right one for well-working bubbles by sonication, without taking into account other suggestions from DST-theory.

After the thorium experiment, we learned how to choose the vessel geometry to obtain the proper energy distribution by following the asymmetry of Lorentz violation [31, 32].

Therefore, we used for nickel experiment this new vessel, quite different from the reaction chamber. In this way, we exploited all the conditions for the DST-reactions: space, time and asymmetry.

From a technological point of view, the two experiments, built around two different types of equipment, varied in the cooling device.

The first one on thorium was equipped with an effective cooling system which allowed an uninterrupted sonication for 90 min.

In the second experiment performed on nickel and described in these pages, the set-up was not equipped with a cooling system, limiting the maximum duration of a run to 150 s (we fixed it at 100 s).

The over-heating due to longer sonication would have damaged the instrument: the sonotrode would have changed its length and the resonance conditions, used to have the adequate energy distributions, would have been lost.

By the way, in order to preserve the integrity of the system, the ultrasound machine was implemented with a security system blocking the electrical supply, thus stopping the machine, if working out of the right parameters.

Notwithstanding the difference between the two experiments, the condition of Local Lorentz Invariance (LLI) violation was achieved in both the cases and the DST-deactivation took place.

In the Nickel experiment, lower power and shorter run-times could be used thanks to the additional knowledge acquired in the meantime.

5 Conclusions

We have presented some technological advances performed in applying the DST-theory to obtain the deactivation of radioactive substances.

In comparison with the previous experiment with thorium, in these years we have been able to understand the role played by the geometry (shape) of the experimental equipment – beside the role played by the energy density and by the size of the micro-reactors (bubbles) – in inducing the LLI breakdown, which is at the basis of the DST-reactions.

Deactivating nickel nitrate ($Ni(NO_3)_2$) made of radioactive Nickel-63 in nitric acid (HNO_3) is a more realistic case, not only because the molecule and the system are more complex but also because the problems related to the highly corrosiveness and to the radioactivity have to be dealt with at the same time, thus mimicking a realistic situation like those of nuclear waste.

In the liquids, the right coupling between the sonotrode and the vessel was a multiple problem to solve. They both must be of the right shape to achieve the conditions of Lorentz violation. In fact, the right energy densities (in space, time and direction) must be reached at the same time to trigger the DST reactions.

By the way, similar multiple problems are faced when producing DST reactions in solids.

To obtain further improvements with respect to the Thorium experiment, ICP-MS (Inductively Coupled Plasma Mass Spectrometry) analyses are in progress [28] to evaluate the reduction of Ni-63, to check if it matches the spectrum reduction and to look for inert nuclei produced by the Ni-63 DST-transformations.

6 Remarks

We are reporting the conclusive experiment from a set, where different vessels and different positions of the sonotrode in the vessel were tested and the corresponding temperature increase was recorded. In this sense, we collected a sound reproducibility of the phenomenon as we verified that the highest temperature increase occurred when the experimental conditions were those predicted by the DST theory to obtain the Lorentz Local Invariance breakdown.

All the precautions required to treat radioactive material in order to prevent and to check contamination and to reduce to the minimum the irradiation (only beta electrons completely screened by water and glass and X-rays) were followed, according to the standards of the International Commission on Radiological Protection (ICRP) recommendations.

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