WATER

Model for Transmutation of Elements using Weak Energy of Water Leading to Faster Disintegration of Radionuclides

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Keywords: Keywords: radionuclide, transmutation of elements, weak energy of water, extended particle, half-life reduction

Received: June 4, 2018; Revised: August 14, 2018; Accepted: October 3, 2018; Published: December 17, 2018; Available Online: December 17, 2018

DOI: 10.14294/2018.7

Abstract

There is great interest among physicists as to whether the transmutation of an element is possible without the use of enormous energy. The topic is also interesting from the viewpoint of basic science. There appears to be reason to suspect that one could change radionuclide transmutation into stable elements with the application of “weak” energy. The discovery of efficient approaches to reduce the half-life of a radionuclide is one of the intriguing issues in current science, and it is also relevant from the perspective of engineering, particularly when nuclear power reactors are decommissioned or demolished. In this context, the purpose of this research is to achieve half-life reduction of radionuclides by using the weak energy of processed water to transform them into stable elements. In terms of practical application, we report the rapid deactivation of cesium (hereafter, Cs) radionuclides (more than 90% reduction in radioactivity) from contaminated soils observed over 5 years after the Fukushima accident. Further, we propose the theoretical idea underlying such transmutation, and we discuss the analytical results leading to the generation of stable barium from radioactive Cs using processed water. Our low-cost and simple approach is suitable for rapidly reducing radioactivity, particularly in the cases of nuclear accidents and decommissioned reactors.

Introduction

Many scientists believe that only a few chemical elements existed at the origin of the earth (as opposed to the variety of elements available today), and they have attempted to create many different elements from one element over a long period of time. Further, scientists are also interested in the possibility of rapidly reducing the radioactivity of nuclear substances. The accepted view in the scientific community is that such reduction is not possible

without the application of methods that use the extremely high energies related to nuclear reactions, and these methods have not been practically employed thus far. In this context, over the past few years, the idea of applying Pd complexes permeated with heavy hydrogen to realize low-energy transmutations has been reported (Iwamura *et al.*, 2002), but suitable mechanisms to transform elements have not thus far been available. Meanwhile, in an earlier work (Kitamura *et al.*, 2009), the absorption of D2 gas by PdZr oxide nanopowder was reported to generate heat and 4He. The goal of the study was to detect the heat and neutrons. In another work (Ohtsuki *et al.*, 2004), the half-life of Be-7 was reduced by 0.83% by encapsulating Be in C60 within a small area with an interatomic distance in the range of nanometers. In all these cases, the methods for transmuting the elements utilized a "dry" system and/or the gaseous phase, or required large amounts of energy. Furthermore, the authors did not present the mechanisms utilized in detail; they only reported on the formation of heat by faster disintegration. In contrast, the system utilized in our work is a "wet" system, i.e., it employs only water. The aim of this research is to reduce the radioactivity of Cs through its faster disintegration, thereby resulting in generation of stable elements; further, the system may be applicable to certain other types of radionuclides generated in nuclear power plants and to radioactive cobalt (half-life of 5.27 years with β-decay similar to Cs) in medical radiation applications. The essential idea in our theoretical consideration relies on elementary particle physics theory (Yukawa, 1950). Our methods and experiments involve the special treatment of common materials, such as water. The high-pressurized water that is used in the treatment forms a unique medium, which we call "Spin Information in Gauge field Network (SIGN)" water in this study. This term represents the hypothetical extended particle that is present in the specially processed water; the particle exists in the quantum state, exhibiting spin and vibration in the manner of electrons and/or protons, thus functioning like an elementary particle. We report a drastic reduction in radioactivity in contaminated soil via the formation of stable elements (focused on element changes of radioactive Cs to stable ones) through the application of SIGN water based on more precise theoretical discussions than those reported in our previous studies (Sugihara, 2013, 2015).

Experimental Methods

The starting material in our method is common tap water, which was processed at high pressures of 110~113 MPa and was maintained for 10 minutes at this pressure during the process of 90 minutes so that the hydrogen bonds were broken. We note that the presence of chemicals in the water does not affect the results and tap water is always used as the control, and the properties of the water are "defined" only after nuclear magnetic resonance (Fourier transform NMR spectrometer R-90H, Hitachi Co. Ltd) analysis and Fourier-transform infrared spectroscopy (FT/IR-6000, JASCO) measurements. This research is focused on the physical state of water rather than its chemical state. Regarding the method, the processed water was added to soil contaminated with the radionuclides of interest (Cs in our case) in identical volumes. The control sample and sample of interest were set apart by approximately 1 m to avoid transfer of energy (or information) from the processed water in an open space. Radioactivity measurements were performed after drying at 333 K in an oven where the sample was directly immersed in a medium (or in a vinyl bag). We used a Geiger–Müller counter (Gamma-Scout Geräte Nr. 043149, Germany) and also a germanium detector (HPGe, GC3019, Meriden, CT, USA) for identifying the energies emitted from Cs-134 and Cs-137. The energies of the two nuclei were

determined using Ge semiconductor detectors. In addition to these instruments, we employed NMR to infer certain characteristics of the water molecules. After the water reacted with the radionuclides, the soil was analyzed to detect the various elements by using inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer 300D), inductively coupled plasma atomic emission spectrometry (ICP-AES, Shimazu ICPS-7510), and X-ray fluorescence spectroscopy (XRF, Shimazu, EDX-700HS).

Theory of Transmutation of Radionuclides

Properties of Specially Processed Water

First, the properties of the water used in the present research are studied to understand how the system works. It has been reported that water usually exhibits different characteristics in certain pressurized processes. While this type of research on water is rare in the literature, one study (Liu, 2000) utilized water at a pressure of one-hundredth (or less) the value required to break hydrogen bonds (required to be more than 0.8 MPa — evaluation is shown later). The characteristics of the specially processed water (hereafter called only "the water") are associated with the creation of certain physical states of water after breakage of hydrogen bonds due to the high pressure, thereby resulting in the generation of a hypothetical particle $<H^+{\sim}e^->$ (neither ion nor atom) that we call an extended particle, which remains stable in this state (plasma-like state). However, the particle cannot be directly observed in the water by using any instrument at present. Only indirect methods can be utilized to study this water, as shown in the Results section (*Figure 1* through *Figure 4*). Our study involves the macroscopic measurement of parameters such as the dielectric constant; the particle oscillates and/or "spins" in the $<H^+{\sim}e^->$ state, and it is dimensionally very small in the stable plasma-like state (less than a few angstroms in size) with its calculated oscillation frequency ranging from approximately 3~10 THz (Sugihara, 2009). Thus, the particle can emit weak electromagnetic waves in the terahertz and far-infrared ranges. The water containing the particle appears to have a tendency to transmit such radiation although terahertz waves are usually absorbed by water (Kawase, 1996). The extended particle $<H^+{\sim}e^->$ behaves as a fermion with spin 1/2 which resonates with the proton inside Cs nucleus when the extended particle has access to it, and a boson with spin 1 (1/2 + 1/2) or 0 (1/2 - 1/2) as H^+ and e^- inside the processed water. Thus, in this water, the hypothetical extended particles may transfer to other substances information such as momentum while vibrating and spinning in the field of long-wavelength radiations. This is the reason why in this study we call the extended particle an "infoton" which was first coined in an earlier work (Sugihara, 2013). Importantly, the extended particle is not similar to a Rydberg atom, in which the electron moves around the nucleus far from the positive ion (similar to an electron in the hydrogen atom). Rydberg atoms can have comparatively large spatial extensions, perhaps more than 1000 times the diameter of "non-Rydberg" atoms. Thus, the extended particle is neither an ion nor in a higher-energy region (than Rydberg atoms), such as a higher-energy region of 13.2 eV ($H \rightarrow H^+$) in Rydberg atoms or 4.3 eV of H-H covalent bond.

Potential and Field Formed by Cs Atom and Extended Particle

We first consider the potential in a theoretical field for the interaction between Cs and the extended particle $<H^+{\sim}e^->$ in the water. Information (i.e., spin and momentum) from the particle can be transferred to the Cs atom under a certain potential and field in a small space, as discussed later (*Figure 5*

and *6*). The medium in our system is water and the information from the particles can be transferred to the Cs nucleus at a lower rate assumed to be 10-15% (~ 5×10^7 m/s) of the rate of light, as shown in calculation later. Another idea that must be considered regarding very small space-time stages is a gauge field existing along with the effect of gravity as per Einstein's equation in such a quantum field, for which case we theoretically predict accelerated disintegration, as proposed in an earlier work (Sugihara, 2015). This idea is rather more important in our case than the Coulomb barrier which involves a long-range force.

Extended Particle Approaching Cs Atoms and Their Interactions in Nucleus

The present study hypothesizes two stages of interaction between the water and cesium according to the distance between them: i) stage I (10^{-9} to 10^{-12} m) and ii) stage II ($<10^{-13}$ m). In stage I, before interaction, the important requirement for the particle to approach the Cs atom is the gravity term in the Einstein's equation including a space distortion (Sugihara, 2015) (rather than the Coulomb barrier ruled by long range force). First, we briefly review the spontaneous β-disintegration of Cs-134 and Cs-137 before elucidation of the nuclear reaction of interest (the Cs nucleus is known to undergo β-disintegration with weak energy). Radioactive Cs-134 emits γ-radiations in the energy range of 563–1365 keV (at 569, 605, 796, and 802 keV) with a half-life of 2.07 years, while Cs-137 emits γ-radiation only at 662 keV with a half-life of 30.3 years (*Figure 7*). The generic equation for β-disintegration in the nucleus can be expressed as ${}^{A}_{Z}Z \rightarrow {}_{Z+1}{}^{A}X' + e^- + \bar{\nu}_e$ (where A represents the mass number and Z the atomic number). Thus, we have ${}^{137}_{55}Cs \rightarrow {}^{137m}_{56}Ba + e^- + \bar{\nu}_e$ in our case. The life of metastable Ba is 26 min, after which it transmutes to ${}^{137}_{56}Ba$, which is a stable element. Next, we present the basic well-known nuclear reactions as below:

$n \rightarrow p + e^- + \bar{\nu}$ (Neutron transforms into proton, electron, and anti-neutrino).

$p \rightarrow n + e^+ + \nu$ (Proton transforms into neutron, positron, and neutrino).

Now, we again consider the case of the extended particle(s) approaching the Cs atom, as illustrated in *Figures 5* and *6*; particles $<H^+ \sim e^->$ may approach and reach the Cs atom in the figure, maintaining their spin (1/2, 1 or 0), while 1/2 inside Cs nucleus. Corresponding to the distance between proton and electron in the infoton $<H^+ \sim e^->$, 1/2+1/2 or 1/2-1/2 may possible, and furthermore, once the extended particle gets into the Cs nucleus, H^+ functions as a fermion of spin 1/2, and there is the case that the electron is caught by an outer orbital of Cs, therefore valence will change after nuclear reaction resulting in ${}^{138}_{56}Ba$ from ${}^{137}_{55}Cs$, so fermion may also exist. Consequently, the abovementioned reactions occur between Cs nucleus and the particles within the system (stage II). This is discussed in terms of energy conservation and mass balance, also briefly referring to the selection rules to satisfy both the Fermi rules and the Gamow–Teller rules for allowed transitions (Fermi, 1953) in the Results and Discussion sections.

Basic Natures of the Water

Firstly, we mention the high pressure to break hydrogen bonds in water to calculate the pressure in aquaporin of membrane protein where water is said to squeeze into the narrowest part of the protein in each living organ at the speed of 3×10^9 /s (Kozono, 2008 and Yausi, 2008) leading to the pressure in region to be 0.75 MPa corresponding to 0.039 eV with simple calculations. The definition of "smallness of water" is shown with NMR spectroscopy indirectly (*Figures 1 and 2*). Almost all SIGN water may easily go through the aquaporin, meanwhile 50~60% of water at the pressure of

one tenth of 100 MPa may go through the protein although there is not quantitative evidence of this. Furthermore, temperature does not increase beyond 353K at most, and the temperature to break the hydrogen bond of water might necessarily be more than 1000K (a simple calculation). As is well known, the temperature of super

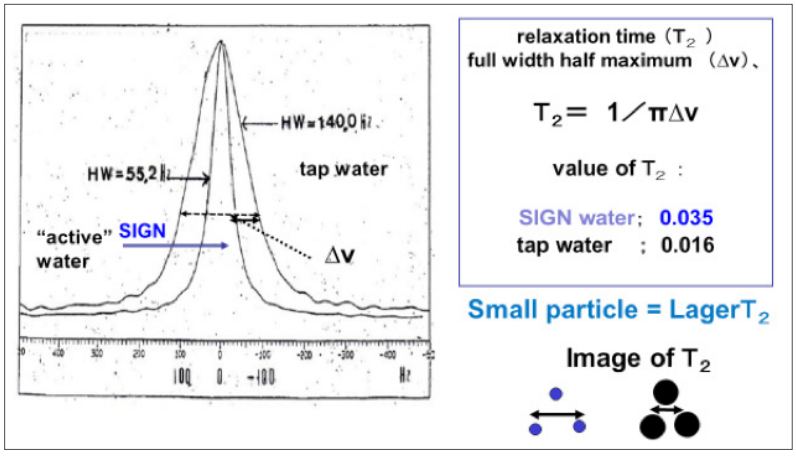
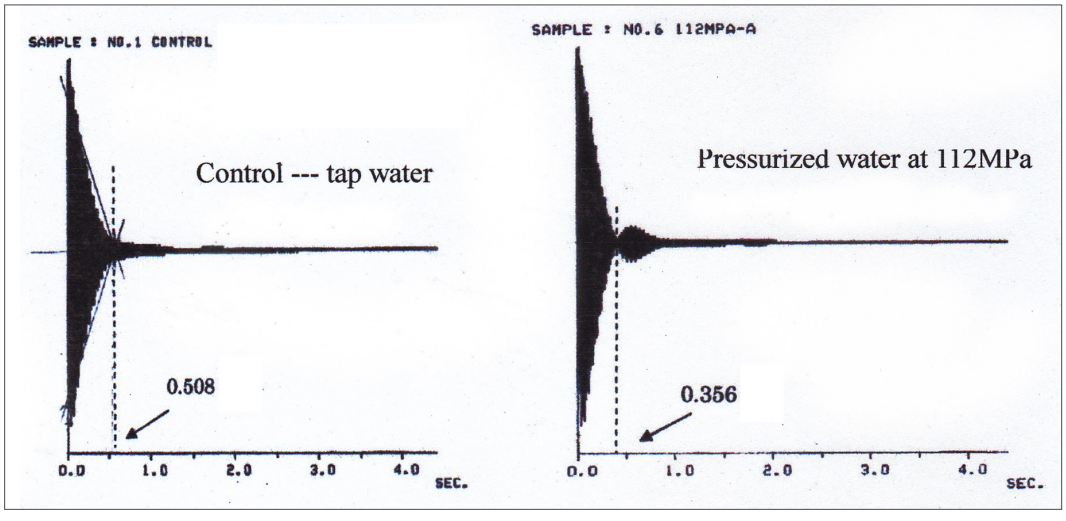


Figure 1. NMR spectroscopy for relaxation time (T2) and free induction decay (FID) for 4 seconds. Upper: T2 and lower: FID.



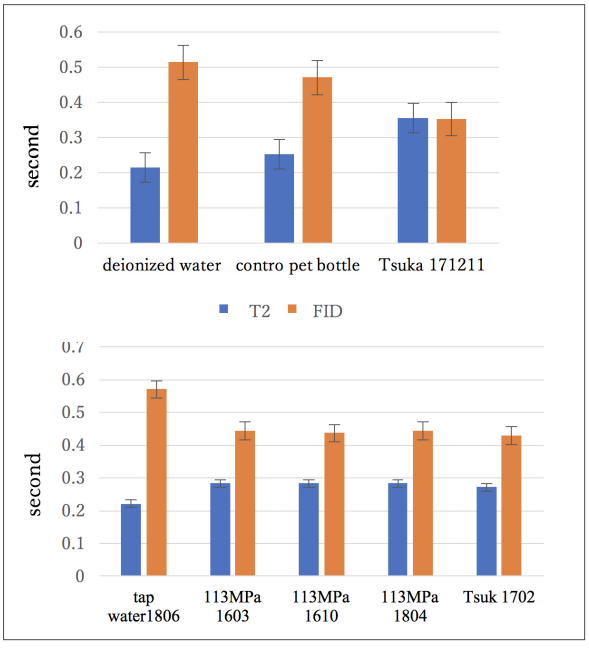


Figure 2. Upper: NMR spectroscopy of both Tsuk 171211 and Tsuk 1702 as compared with deionized water and control pet bottle water. Lower: stability of SIGN water (Tsuk 1702) along with an elapsed time (2016/3 ~ 2018/4).

critical water is said to be 620K although its pressure is about 22 MPa.

After breakage of hydrogen bonds, the water was analyzed by means of an NMR spectrometer as shown in *Figure 1* and some results of NMR in *Figure 2* being followed by terahertz spectrometer (*Figure 3*), and FTIR spectrometer (*Figure 4*).

We can know that T2 is larger for the pressurized water, meanwhile FID is smaller for the processed water. *Figure 2* shows the results of NMR data of the water as compared with other water such as deionized water and pet bottle water sold at a shop as control. Remarkably we can know larger T2 and smaller FID for the processed water (Tsuk 171211 and 1702).

Furthermore, it is shown that the water has not changed with the elapsed time in the lower part of *Figure 2*.

Figure 3 indicated the NMR results of T2 and FID as well as smallness of the water vs pressure.

From the view of electromagnetic wave, FTIR spectroscopies are introduced focusing on the lowest energy of infrared spectroscopy in *Figure 5* as well.

THz wave is generally absorbed by water, but the processed water indicates a little more transmittance. Furthermore, we analyzed an isotope of heavy hydrogen (^{2}H) in

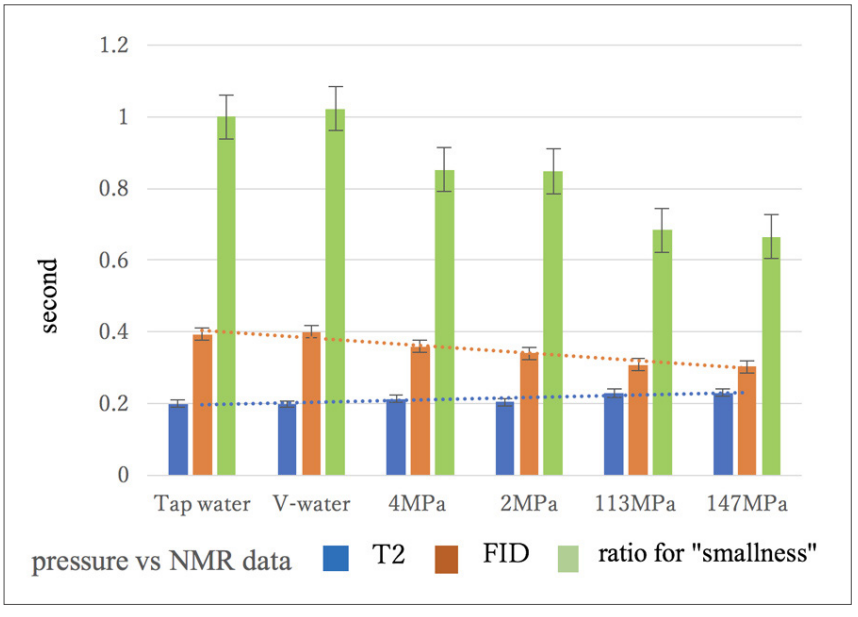


Figure 3. Estimation for smallness of water as well as NMR data (T2 and FID) against each pressure, presenting smallness of the water which is defined in the equation FID/T2.

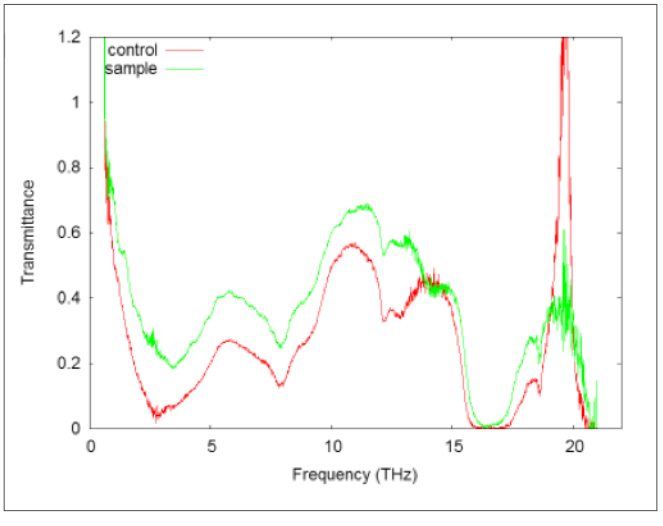


Figure 4. The spectroscopy in THz region. Abscissa shows THz frequencies, and vertical axis shows transmittance. Sample (green); SIGN water, Control (red); tap water.

water. The ratio between ^{2}H and ^{1}H given by the equation:

$$\delta^{2}H = \{(^{2}H / {}^{1}H)_{\text{sample}} / (^{2}H / {}^{1}H)_{\text{standard}} - 1\} \times 100$$

δ is defined by this formula as the ratio of heavy hydrogen (containing a neutron) and hydrogen in the sample compared with that in the standard (control). As the results, the values of δ2H was -58.72 and -65.20 for tap water and the processed water, respectively.

We also confirmed via NMR spectrometry that these particles exist stably over time. Furthermore, in terms of an energy, i.e. the configuration has not necessarily been fixed arbitrarily, and the number of O–H atom configurations in ice is defined as the term W corresponding to redundant entropy which appears when the structure of water is not always formed in the fixed atomic configuration. Thus, we have the redundant entropy of ice, k ln W = 0.806 kcal/mol (Pauling, 1960), which is equivalent to 0.035 eV (8.5

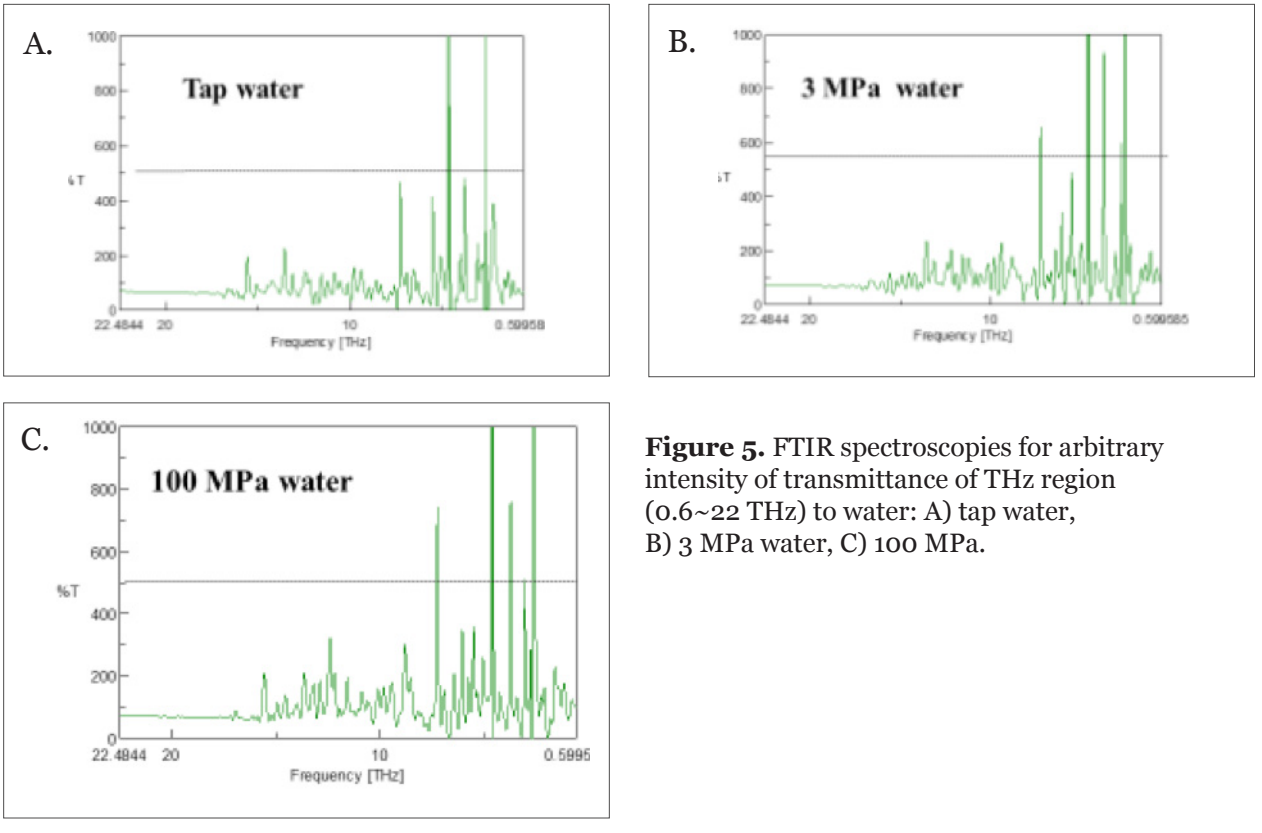


Figure 5. FTIR spectroscopies for arbitrary intensity of transmittance of THz region (0.6~22 THz) to water: A) tap water, B) 3 MPa water, C) 100 MPa.

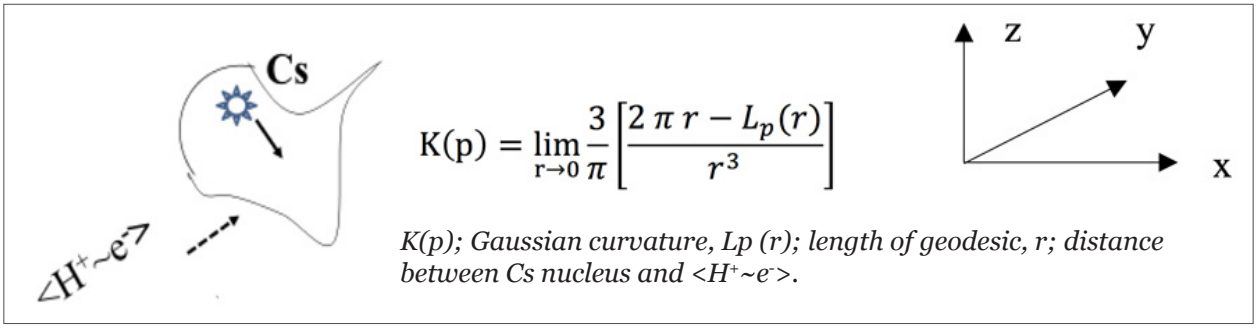


Figure 6. Idea of the potential field between Cs nucleus and particle, showing Gaussian curvature defined as a pommel-shaped potential curve on the coordinates (x, y, z), where Cs nucleus exists higher potential and <H⁺~e> in lower.

THz), where entropy, S = k ln W + U/T (k: Boltzmann constant, W; the number of the state, U; internal energy, which is a function of probability of changing from the states A to B, and the first term corresponds to redundant entropy which appears as the structure of water is not always formed in the fixed atomic configuration.

Applications of the Water to Cs Nucleus

For application to our processed water system, with a configuration possibility of one hydrogen atom in two water molecules, *W* is calculated to be 1.5 as abovementioned, where *k* denoting the Boltzmann constant, and *K(p)* is Gaussian curvature and *p* is momentum.

Figure 6 illustrates the potential field formed by the Cs nucleus and the particle <H^+~e^-> is going upward while Cs moves downward.

The extended particle may possess a larger geometrical cross section of the reaction as compared with the ordinary water molecule as shown in *Figure 7* and detail discussion is later. The hydrogen bond in two cases is cited at approximately 100 pm from the nucleus, and the two sets of dipoles function as a quadruple state. The angles are geometrically calculated in the cesium crystal. Thus, the particles traverse the field at a distance on the order of picometers to absorb a certain amount of the γ-ray photon energies from Cs-137 (662 keV) and Cs-134 (563–1365 keV, as listed previously) in the electromagnetic field and gauge field. Furthermore, the particle can experience acceleration upon approaching the Cs atom, as elucidated in the Discussion section. When two extended particles approach Cs, they may generate a quadruple state leading to spin–spin interactions among smaller particles of the form <H^+~e^->, when compared with the ordinary water molecule. Finally, the particle may reach the Cs atom at 10-15% of the speed of light, corresponding to an energy of 9.2 MeV that is slightly higher than the nuclear binding energy of Cs-137 (approximately 8.5 MeV). Therefore, the particle can enter the Cs nucleus, which comprises stage II. Once the extended particles enter the cesium nucleus, a nuclear reaction occurs. This corresponds to the experimental finding of certain stable

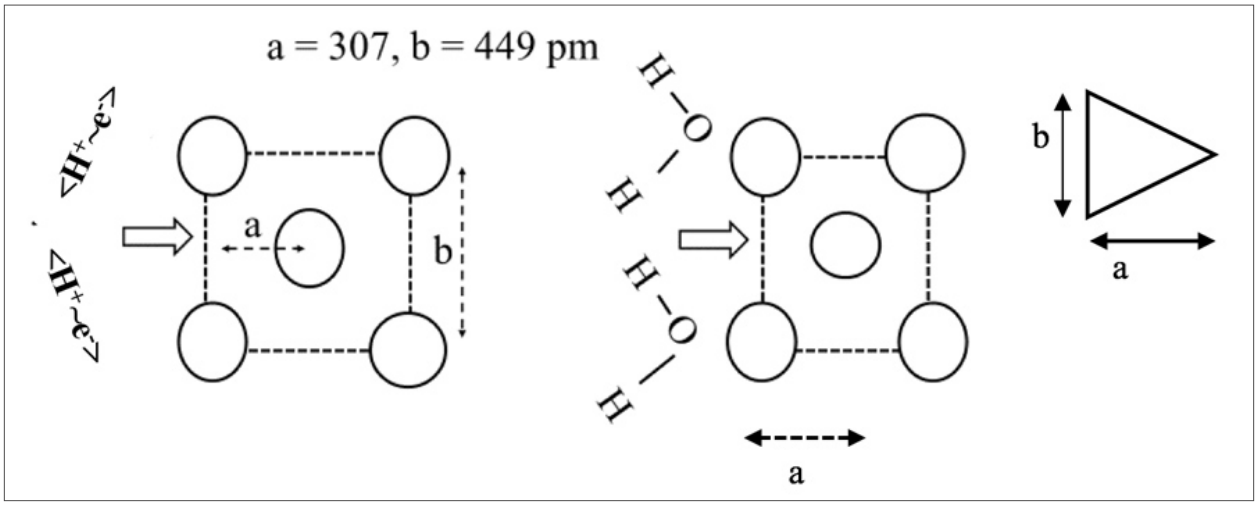


Figure 7. Estimation of geometrical cross section* in the cases of the extended particle (left) and ordinary water molecule (right) approaching Cs nucleus along the direction indicated by the solid arrow.

* Cs crystal and extended particle
tan θ = b/a = 224.5/307 = 0.731
θ = 36.2 (sin2θ = 0.349)

Cs crystal and water molecule
tan θ = b/a = 224.5/897 = 0.250
θ = 14.5 (sin2θ = 0.063)

elements (as indicated by the spontaneous disintegration mentioned in a later section) in the soil processed with the water, thereby leading to a drastic reduction in radioactivity.

In this study, we determined the energy conservation during the nuclear reaction and material balances via calculations (the details are provided in the Discussion section). The calculations indicated that basic β-disintegration reactions, such as, $n \rightarrow p + e^- + \bar{\nu}$ and $p \rightarrow n + e^+ + \nu$ are closely related to the interaction with the extended particle(s).

One of the interesting features observed was that the mass of the particle was closer to that of a neutron than that of a proton, and these particles might be considered to function as both fermions and bosons. There are two quantum numbers, such as, total momentum and spin. The spin of proton and neutron in Cs nucleus are 1/2, respectively.

The spontaneous decay of cesium 134 and

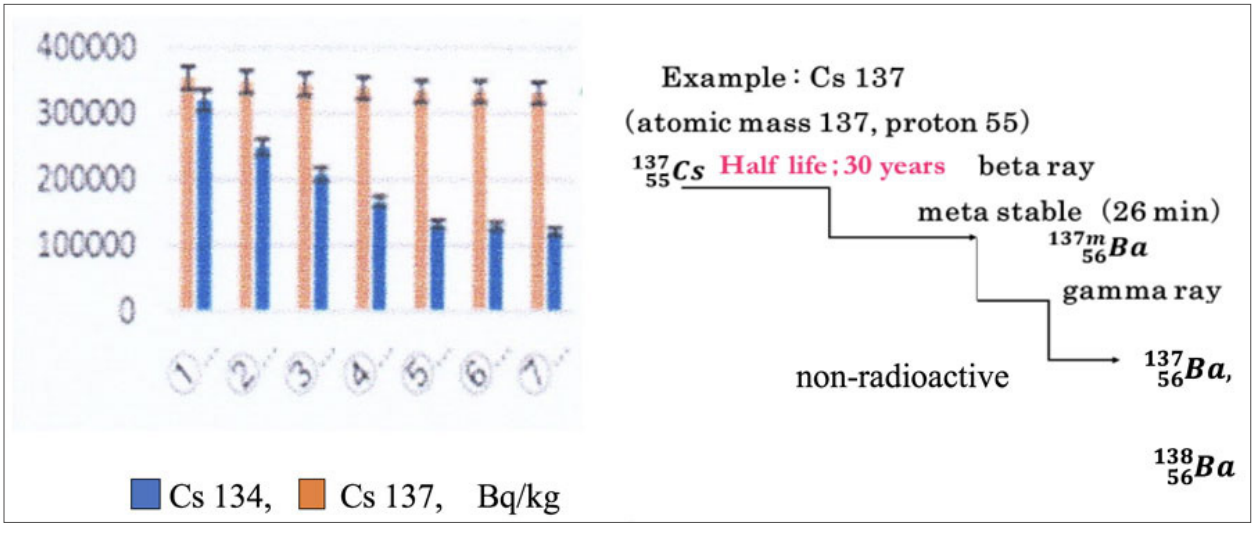


Figure 8. Left; the spontaneous decay of Cs-134 and Cs-137. (1) corresponds to 2011/6/29 and followed by (7) corresponding to 2014/6/10 in 3 years according to Figure 8. Right; changes to Ba from Cs-137 in a spontaneous decay.

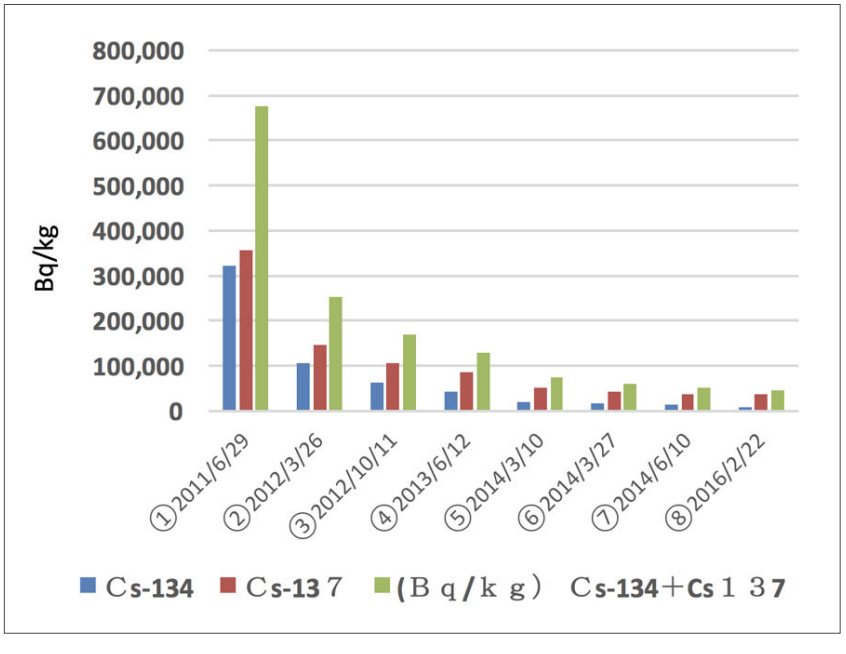


Figure 9.
Changes over five years following residual radionuclides. The results were tracked during the five years after the Fukushima nuclear power plant accident.

137 is depicted in *Figure 8*. We also experimentally determined the changes in the radioactivity of the contaminated soil from the Fukushima nuclear accident (radioactivity values listed for five years after the accident as shown in *Figure 9*.

According to *Figure 8* (right), only half of Cs-137 has changed to non-radioactive Ba after 30 years, and a half of Cs-134 changed to non-radioactive Ba-135 after 2 years spontaneously as shown in the *Figure 8*, left (4) which corresponds to the date of 2013/6/12.

Now, we calculated the amount of Ba atoms generated from radioactive Cs qualitatively. The upper graph of *Figure 10* indicates the number of atoms of Cs-134 and Cs-137 calculated from Bq/kg of actual measured values along with the elapsed time for one year.

Note:

1) The usual chemical analysis (ICP-MS) of Ba is reported for Ba-137, although stable Ba-135 is generated from Cs-134 and stable Ba-138 from Cs-137.

2) The values obtained in May 2011 were obtained just after sampling of the contaminated soils, and subsequently, decontamination was carried out with the water.

Figure 10 shows the decreasing radioactivity of the contaminated soils with SIGN water (processed water) increasing of the amount of Ba atoms.

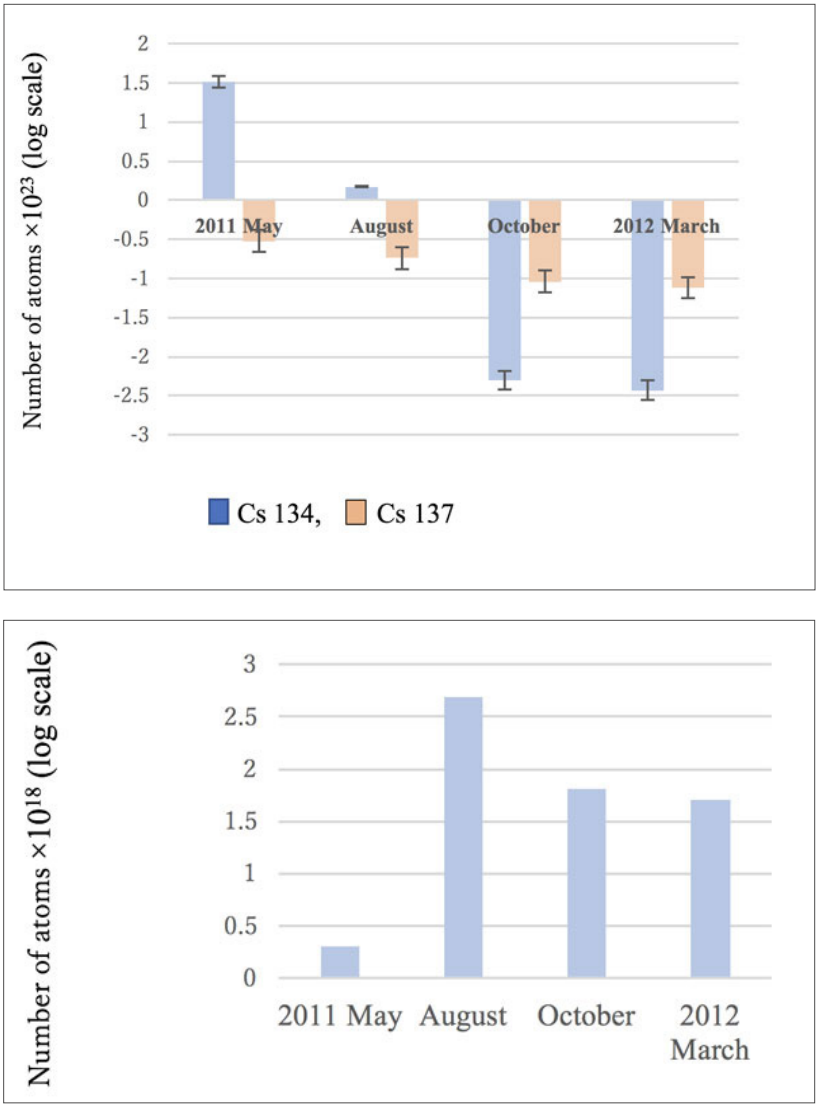


Figure 10. Upper; number of atoms of Cs-134 and Cs-137 calculated from Bq/kg along with the elapsed time, Lower; number of Ba atoms generated from Cs nuclides by reaction with the processed water.

Discussion

Basic Natures of the Processed Water

The processed water can be identified as special water from the viewpoints of electromagnetic wave and the particle. Firstly, we introduced the water after hydrogen bond breaking for which we have proposed to evaluate the status with NMR, THz and FTIR spectroscopies as shown in *Figures 1, 2, 3, 4* and *5*, respectively. And we may thus conclude that we have obtained smaller water when T2 is larger and FID is smaller, and we may get SIGN water having the information generated with the particle after breaking hydrogen bonds. We also employed FTIR in THz region (*Figure 5*) and we can judge that the water transmits THz waves more efficiently than normal water. The water containing the extended particles possesses unique properties. First, the particle emits long-wavelength electromagnetic waves due to vibration and spinning, which we may suppose acts like a plasma-like state from the dielectric constant in the equation of plasma oscillation, $\omega^2 = \frac{ne^2}{m\epsilon} \cong$ 5THz, where n of water molecule = 2.7×10^{-25}/m3, e; constant=1.6×10^{-19} C, m; $H^+ + e^- = 1.67 \times 10^{-27}$ kg, $\varepsilon \approx 80$ (Malmberg, et.,al. 1956).

We can consider the water to be the particle leading to an element change. As analyzed in the values of δ2H, SIGN water had a larger δ2H (-65.20) than tap water (-58.72). It may not necessarily mean increment of heavy hydrogen; the isotope analysis is based on the mass of heavy hydrogen which involves a neutron, then the mass (H^++ neutron) increases. However, the mass of the extended particle <H^+~e^-> in the processed water closes to neutron (difference is only 0.08%). Therefore, δ^2H becomes larger, measured as increase of heavy hydrogen in the water. In addition to mass of the particle (nearly equal to that of a neutron), quantum number of spin which can be expressed in two states, such as fermion 1/2, and boson with spin 1 (1/2+1/2) or 0 (1/2−1/2) corresponds to the distance between the proton and the electron in the extended particle. Once the particle gets into the Cs nucleus, the particle behaves as a neutron with spin 1/2. There is the case that the electron (spin, 1/2) is caught by an outer orbital of Cs, therefore valence will change after nuclear reaction resulting in $^{138}_{56}Ba$ from $^{137}_{55}Cs$. Thus, the nuclear spin value, ΔI = 0 or ΔI = 1, may satisfy the criterion for the selection rules in the water before the reaction due to the particle's physical state that is neither atom nor ion. Now, the spin of the particle <H^+~e^-> is considered to have two values, such as, ΔI = 0 (1/2 − 1/2) or ΔI = 1(1/2 + 1/2), characterized by the state being not only H^+ and not only electron controlled by the momentum and the coordinate. In the transition of β-disintegration, the parameters controlled by the selection rules are total angular momentum and parity. I presumed in the water, ΔI = 0 when H^+ and e^- are close and ΔI = 1 when they are far.

The mathematical development like Heisenberg representations should be discussed in order to elucidate the selection rules, but it does not seem to fall within the scope of our current discussion.

Interaction of the Processed Water with Cs

In the system of radioactive Cs and the water, the presence of a pommel-shaped potential relating to Gaussian curvature (*Figure 6)* appears to aid the nuclear reaction between the particle and the Cs atom, wherein the particle can access and enter into the nucleus as shown in *Figure 7*. The required energy for overcoming the Coulomb barrier can be evaluated as follows: The particle (electrical charge: e, mass: m) emits energy (E)—information—per unit time over a solid angle dΩ generated by acceleration (a), angle (θ) (defined in *Figure 7*) and c for speed of light as indicated in the following equation:

$$E = \frac{(ae)^2}{c^3} sin^2\theta \frac{d\Omega}{4\pi} \quad (1)$$

Energy E increases with θ being vertical with respect to the x-axis; the axes of the x–y plane form θ or the axes of the y–z plane form θ in the case of spatial separation corresponding to stage I (between cesium and the extended particle), thereby indicating that the curvature of the potential becomes larger toward the cesium atom, and the particles can also obtain energy from Cs nucleus. The extended particles or water molecules approach the Cs crystal with a geometrical cross section as described in *Figure 7*. As a result, a nuclear reaction may occur at room temperature without the need for large amounts of external energy.

Next, we discuss the aspects of energy conservation and material balances. Once the extended particles enter the Cs nucleus, nuclear reactions may occur between them. As mentioned previously, a drastic reduction in the radioactivity (in the soil from the Fukushima accident) was observed, and thereafter, certain stable elements were experimentally observed in the soil processed with the water. The calculation of the energy conservation is as follows for the reaction of electron emission: $n \rightarrow p + e^- + \bar{\nu}$ (neutron transforms into proton, electron, and anti-neutrino). The masses of the neutron, proton, electron, and the extended particle can be calculated as m_n (1.00898 u), m_p (1.00759 u), m_e (0.000548 u), and m_{ex} (1.008138 u = 1.00759 u + 0.000548 u), respectively. The reason as to why the extended particle $<H^+ \sim e^->$ is assigned mass m_{ex} (1.008138 u) instead of the neutron mass can be explained as follows; the mass relation, m_n (1.00898 u) > m_p (1.00759 u), which corresponds to a mass difference of 0.14%. In considering the mass of the extended particle $<H^+ \sim e^->$ instead of that of the neutron, the particle will possess mass m_{ex} (1.008138 u) closer to that of the neutron (m_n = 1.00898 u, a difference of only 0.08%) as shown previously, and its spin is similar to that of the neutron as well. When the neutron transforms into a proton and certain other elementary particle in the electron emission during β-disintegration, the extended particle takes the place of the neutron as shown above. The mass increases, adding to that of a proton existing already in the right-hand side due to the presence of H^+ in the extended particle $<H^+ \sim e^->$ on the left-hand side. Therefore, the system mass increases, leading to a change in A_{Cs}+1, corresponding to an element, such as $^{138}_{56}Ba$; $^{A}_{Z}A_{Cs}$ and $^{A+1}_{Z+1}A_{Ba}$ indicate the cesium atom and barium, respectively:

$$n \rightarrow p + e^- + \bar{\nu} \quad (p + e^- = A_{<ex>} + A_{Cs} = A_{<ex>} + A_{Ba})$$

$$^{137}Cs + <H^+ \sim e^-> \ (\simeq n) \rightarrow {}^{138}Ba + e^- + \nu$$

The following equations show positron emission during β-disintegration. The $<H^+ \sim e^->$ takes the place of the neutron in the right-hand side, and thus the mass does not change as following; A_{ex} indicates the atom of the extended particle, A_{Ba}, barium, and so on.

$$p \rightarrow n + e^+ + \upsilon \ (A_{<ex>} + A_{Cs} = A_{<ex>} + A_{Ba})$$

$$^{137}Cs + <H^+ \sim e^-> \rightarrow n \ (\simeq H^+ \sim e^->) + {}^{137}Ba + e^+ + \nu.$$

Next, the energy balances are calculated as follows: Let us consider the energy conservation of the reaction in the electron emission using the following equation (Halliday 1955).

$$E1 + E2 = MV^2 \quad (2)$$

Two reactions are considered as follows: E_1 and E_2 represent the total energies of two of

the elementary particles (m_n and m_p, m_{ex} and m_p, etc.) First, we calculate the total energy ($E_1 + E_2$) for m_n (=1.00898 u), m_p (=100759 u), and m_e (=0.000548 u) using *Equation (2)*. Here, V represents the speed of the extended particle in the water, which we set as 10–15% of the speed of light in vacuum to be conservative. Only 5% the speed of light is needed to achieve 1.18 MeV of kinetic energy of the particle although this has not been justified experimentally and is discussed more later. A similar calculation can be applied to the case of positron emission as following,

$$m_n V^2 - (E_1 + E_2) = 0 \qquad (3)$$

Upon replacing the neutron with the extended particle, we note from the calculation of energy conservation for electron emission that there is no energy difference in the reaction, i.e., $m_{ex}V^2 - (E1 + E2) = 0$; on the other hand, the energy difference in the case of positron emission is 1.35 keV. Thus, the extended particle in the electron emission appears to easily react with the nucleus of cesium in comparison with the case of positron emission. The results of this evaluation were in agreement with the theoretical analysis in which Ba-138 (69%) is formed to a greater extent than Ba-137 (31%) (Sugihara, 2013). Next, we examine the detailed energy calculations below. First, the energy balance is calculated for electron emission: $n \rightarrow p + e^- + \bar{\nu}$ (neutron transforms into proton, electron, and anti-neutrino). The masses of the neutron, proton, electron, and the extended particle are set as m_n (1.00898 u), m_p (100759 u), me (0.000548 u), and m_{ex} (1.008138 u = 1.00759 u + 0.000548 u), respectively. The reasons as to why the extended particle $<H^+ \sim e^->$ is considered instead of the neutron have been discussed previously. Thus, we have

$$E_1 = \frac{(m_{ex}^2 + m_p^2 - m_e^2)V^2}{2\,m_{ex}} = \frac{(1.00814^2 + 1.00759^2 - 0.000548^2)(2.9979\times10^8\times0.75)^2}{2\times1.00814} = 5.093787\times10^{16}$$

$$E_2 = \frac{(m_{ex}^2 + m_e^2 - m_p^2)V^2}{2\,m_{ex}} = \frac{(1.00814^2 + 0.000548^2 - 1.00759^2)(2.9979\times10^8\times0.75)^2}{2\times1.00814} = -0.002778\times10^{16}$$

and E1 + E2 = 5.09657×10^{16}, while $m_{ex}V^2$ on the right-hand side is calculated to be $1.00814 \times (2.9979 \times 10^8 \times 0.75)^2 = 5.09657 \times 10^{16}$, i.e.,

$$\Delta E = m_{ex}V^2 - (E1 + E2) = 0 \qquad (4)$$

Now, the extended particle $<H^+ \sim e^->$ follows the positron emission process described below: $p \longrightarrow n + e^+ + \nu$ (proton transforms into neutron, positron, and neutrino). Thus, we have

$$E_1 = \frac{(m_p^2 + m_{ex}^2 - m_e^2)V^2}{2\,m_p} = \frac{(1.00759^2 + 1.00814^2 - 0.000548^2)(2.9979\times10^8\times0.75)^2}{2\times1.00759} = 2.26673\times10^{16}$$

$$E_2 = \frac{(m_p^2 + m_e^2 - m_{ex}^2)V^2}{2\,m_p} = \frac{(1.00759^2 + 0.000548^2 - 1.00814^2)(2.9979\times10^8\times0.75)^2}{2\times1.00759} = -0.00124\times10^{16}$$

and E1 + E2 = 2.26549×10^{16}, while $m_p v2$ on the right-hand side is calculated to be $1.00759 \times (2.9979 \times 10^8 \times 0.75)^2$, i.e.,

$$\Delta E = m_p V^2 - (E1 + E2) = 1 \times 10^{-6} \text{ (corresponding to 1.35 keV)} \qquad (5)$$

Therefore, electron emission is easier than positron emission, because the energy differences before and after the reaction are 0 keV for electron emission and 1.35 keV for positron emission according to Equations (4) and (5), respectively.

Furthermore, we analytically found the stable elements formed by the reaction of radioactive cesium with the water. Here, we mention that other stable elements, such as, lanthanum and cerium were also found in concentrations greater than those usually found in soil.

Next, we discuss the formation of stable elements via the abovementioned reactions according to the lower part of *Figure 10*. We can compare with the amount of Ba-137 normally observed in the ground, which is $2.2 \times 10^{15\text{-}16}$/mol which is the larger amount of Ba generated from Cs in our system, 4.9×10^{18}/mol. Ba was reduced to 31.9 (October 2011) from 128.9 × 104 Bq/kg (May 2011), which was a reduction of more than 75% over 5 months. As discussed previously, the corresponding reaction was posited as ${}^{137}_{55}Cs$ + <H^+~e^-> → ${}^{138}_{56}Ba$. The concentrations of Ba, La, and Ce were found to be Ba = 63.3%, La = 21.3%, and Ce = 15.4%, which were close to the corresponding theoretically obtained values of Ba = 52%, La = 32%, and Ce = 16% from the application of group theory to the cesium/extended-particle system. One of the reasons for this drastic reduction in radioactivity (*Figure 9*) was the rapid change of Cs-134 and Cs-137 into Ba, La, and Ce when using the water.

Conclusions

From the findings of our study on the application of the processed water to radionuclide reduction, we draw the following conclusions:

1) Processed water (resulting from only the dissociation of hydrogen bonds), named SIGN water, was introduced as a novel material to generate stable elements such as barium, lanthanum, and cerium from treated radionuclide-contaminated soil.

2) A drastic reduction in radioactivity was observed, and this reduction mechanism was discussed from the theoretical and experimental points of view based on β-disintegration of weak energy.

3) This method can be applied to reduce large volumes of radioactive substances, and it is inexpensive, simple, and fast, except in the cases of heavier nuclei such as uranium and plutonium. However, any size of our advanced technology system can be installed corresponding to the scale of the facility with the existing engineering infrastructure relating to nuclear materials and/or waste.

4) We are continuing our research into quantum mechanical behavior of element changes by the water, after breaking H-bonds.

Discussion with Reviewers

Reviewer: It is difficult to deactivate heavier nuclei such as uranium and plutonium using the method proposed in this study because these elements undergo nuclear fission for transmutation, and they must be treated with high energy before deactivation. Why? This is a very interesting point, please elaborate, explain and cite properly.

Author: Basically, heavier nuclei (radioactive ones) decay spontaneously into certain stable elements by nuclear fission, emitting radioactive radiation such as α-ray (${}^4_2He$)He particle) to decrease it's mass for stable state, which is called a spontaneous decay. For example, ${}^{226}_{88}Ra$ → α decay → ${}^{222}_{86}Rn$ (radon) +γ, and ${}^{227}_{88}Ra$, ${}^{227}_{88}Ra$, ${}^{230}_{88}Ra$. However, lighter elements like Cs emit β-ray (electron) and γ-ray (photon) for increasing stability. According to author's experience, the elements under 88 atomic number may

become stable elements by nuclear fusion (not nuclear fission) although this has not been established evidently.

Reviewer: Please give the exact pressure, time and (average) composition of the tap water used (even if the chemical composition is less relevant as the author later explains).

Author: The tap water was kept at 10 min. to 113 MPa in the process lasting 90 min. The chemical analysis data (ppm) of the tap water, Na: 1100, Mg: 26000, Ca: 18000, Sr: 1100, S: 630, Al: 56, Si: 22, K: 20, Cr: 38, Mn: 23, Fe: 22, Co: 11, Zn: 34, Y: 13, Cd: 10, Cs: 15, Ba: 24 and less than 10 ppm: Li, B, P, Ti, Ni, Cu, As, Zr, Nb, Mo, Rh, Pd, Ag, La, Ce, W and Pb (by ICP-AES instrument). By the way, Cs isotope in this analysis was the stable one: $^{133}_{55}Cs$.

Reviewer: The author states that the speed of the particle <H^+~e^-> is assumed to be about 10-15% of the speed of light. How is this important assumption justified? This is a crucial point to overcome the Coulomb barrier.

Author: I assumed that "about 10-15% of the light speed" is just conservative. Even just 5% of light speed is 1.18 MeV kinetic energy of the particle, although, unfortunately, this is not justified. However, similar parameters, like mass, are near to a neutron compared with proton (discussed in the manuscript), and Cs 137 decay mode β-(1.177 MeV) γ is almost the same as the kinetic energy of the particle at 5% light speed. Furthermore, the particles obtaining the energy from Cs leads to higher momentum (P=mv; m, mass and v, velocity) so that they may overcome the Coulomb barrier.

Reviewer: Due to the crucial role of the high pressure processed water, the author should further discuss the water state so obtained.

Author: In the article, the author added a sentence for elucidating the basic nature of water regarding electromagnetic waves. Namely, the changing of the angle in H-O-H is said to exist around a far-infrared region, although SIGN water may also emit far IR, close to the frequency of THz (3~10 eV). Therefore, the processed water possesses an altered angle due to the particle <H^+~e^-> and the THz wave vibrates perpendicularly, going with progressing direction, which results in higher transmittance. Meanwhile, <H^+~e^-> may get into the Cs nucleus as a particle. The author added the data including NMR spectroscopy that somewhat corroborate this assumption.

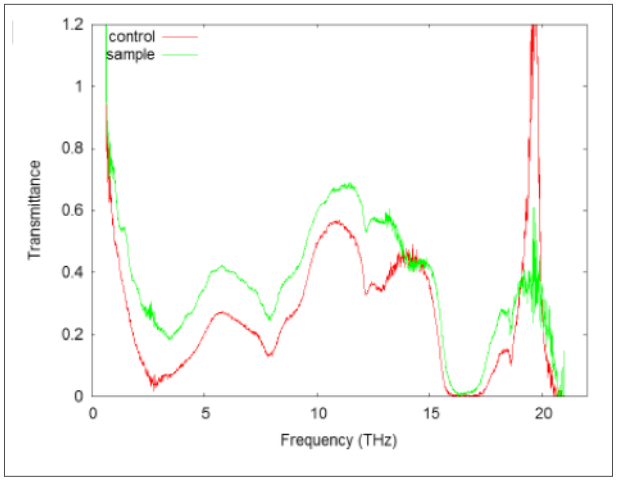
Reviewer: The author states that hypothetical extended particles <H^+~e^-> are formed, and they are neither ions nor atoms and cannot be directly observed in water having small dimensions, and behave as a stable plasma-like state with oscillation frequency ranging from 3-10 THz. How does the author know all this? He says the water dielectric constant is affected by the presence of these particles in water. Which ones are the quantitative changes of the water dielectric constant? "The water containing the particle appears to have a tendency to transmit such radiation although terahertz waves are usually absorbed by water." Please, explain this point. How is it possible?

Author: I put down a precise calculation in the following:

The particle emits long-wavelength electromagnetic waves due to vibration and spinning, which we may suppose plasma-like state from dielectric constant in the equation of plasma oscillation,

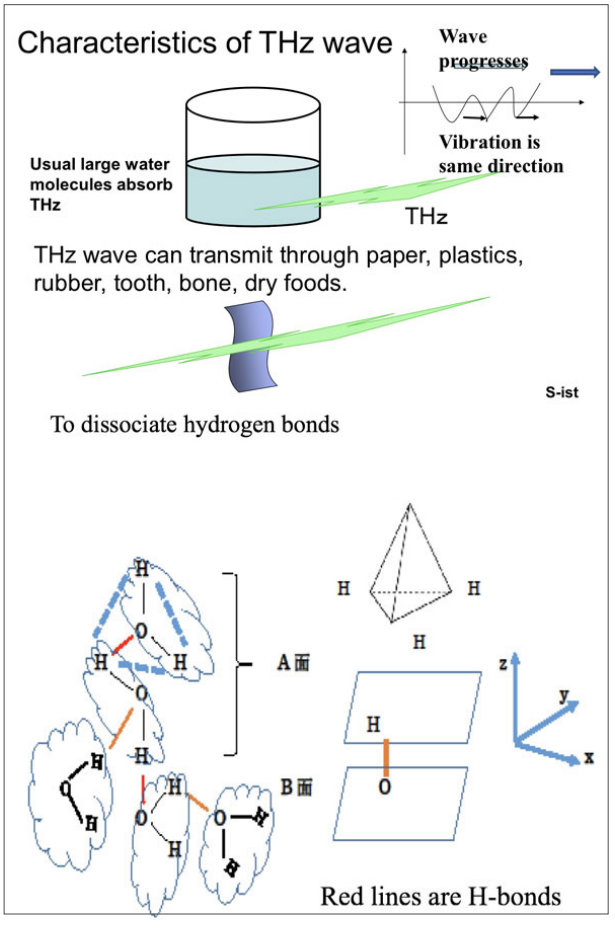
$\omega^2 = \frac{ne^2}{m\epsilon} \cong$ 5THz, where n of water molecule=$2.7 \times 10^{25}/m^3$, e; constant=1.6×10^{-19}C, m; $H^+ + e^- = 1.67\times 10^{-27}$ kg, $\varepsilon\approx 80$. *)

*Malmberg, C. G. *et al., Journal of Research of the National Bureau of Standards* Vol. 56, No. I, January 1956.



Appendix Figure 1: THz spectroscopy of water (measured at Tohoku Univ. Sendai, 2007). Appendix Figure 1 shows the evidence connecting the THz region with SIGN water. Water generally adsorbs THz waves, but SIGN water indicates a little more transmittance.

control: tap water, sample: SIGN water.



Appendix Figure 2: Characteristics of THz wave (top) and to dissociate hydrogen bonds (bottom).

Considering the effect on dielectric constant of the presence of the particles, only date was published in *Water* Vol. 1 pp. 92-99 (Sugihara, 2009) when the constant was measured at less than 1 THz resulting in a difference of $\varepsilon \propto 4$.

Now, considering this point, "have a tendency to transmit such radiation although terahertz waves are usually absorbed by water," see *Appendix Fig. 1.*

A small water molecule may change bond angles as shown in *Appendix Fig. 2.*

Reviewer: Microorganisms may be activated by the activated water, and they may enhance the transmutation of radioactive elements. No consideration of this possibility is found in the manuscript.

Author: Yes, I agree about the possibility of the existing microorganisms and that they may have an effect on the reduction of radioactivity. I came to this idea through an experiment after the submission of this manuscript to WATER Journal. So, I will discuss the quantum mechanical behavior of elements' transmutation connected to microorganisms in the future.

Acknowledgments

I thank the senator/farmer of Nihonmatsu city, Mr. G. Satoh, for providing a warehouse and rice field for large-scale testing, and also Mr. Y. Nagasaka (Brighten Inc.) and Mr. N. Iwamoto (T.I.C. Inc.) for sampling and measurements. I express my gratitude to Dr. Murata (Coallite Inc.) for discussions and testing of the reduction in radioactivity in the contaminated soil in Fukushima.

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