Research Article

Destruction of Radioactivity by Stimulation of Nuclear Transmutation Reactions

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Abstract

Modern science considers properties of radioactive decay as intrinsic characteristics of each isotope that cannot be affected by changes in the surroundings. Here, we present an approach for stimulation of nuclear transmutation reactions leading to accelerated destruction of radioactive isotopes that allows expedited disposal of radioactive materials.
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1. Introduction

The disposal of radioactive materials is one of the most important tasks to be resolved by the humankind. This process can be started only after radioactivity has decayed enough to allow further utilization via decontamination of the radioactive materials and subsequent return of the decontaminated materials and removed radioactive isotopes for a second use and/or storage. Therefore, accelerating the reduction of radioactivity is an economically viable option of radioactive waste treatment in the nuclear industry.

2. Background

In theory, accelerating the reduction of waste radioactivity is possible through transmutation nuclear reactions of the radioactive isotopes present in the waste by stimulating the radioactive decay of the isotopes and/or nuclear reactions of the radioactive isotopes with other isotopes to form stable isotopes. Transmutation of any radioactive isotopes leading to the formation of stable isotopes when no radioactivity is produced results in destruction of radioactivity. However, it is necessary not just to prove that it is possible but also to find a stimulus acceptable from practical view to accelerate the reduction of waste radioactivity.

Modern science considers rates of radioactive decay as intrinsic characteristics of each isotope that cannot be affected by changes in the surroundings. The radioactive decay of an isotope is measured in terms of "half-life," the duration

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of a period necessary to reduce the radioactivity of the isotope by half. The half-life values of the most common decay processes are well known. However, an analysis of published scientific literature shows that certain direct and indirect experimental data challenge this approach.

One of the first methods of changing the half-life of isotopes [1] involved heating an isotope to a very high temperature (2E+8 K), which caused a part of the ^{176}Lu to decay to ^{176}Hf by passing the conventional slow route, and going into an isomeric state with a half-life of only 3.68 h. Another approach was patented by Backer [2]. He suggested that applying electrical potential of 50–500 kV to contaminated metals that contain alpha, beta, or gamma radioactivity and have been placed as a target in a Van de Graph accelerator would significantly accelerate decay. It was shown that the decay for alpha emitter, thorium 230 ($_{90}$Th^{230}), can be accelerated by a factor of 4.49×10^4 while for beta minus emitter, thallium 204 ($_{81}$Tl^{204}), a respective acceleration factor of just 15 was measured. This significant difference can be explained by the fact that alpha decay process is controlled by the Coulomb barrier, which was modified by applying electrical potential, while beta decay is controlled by electron–nucleus contact. Jung et al. [3] found conditions to transmute even a stable atom of ^{163}Ds$_{66}$ to ^{163}Ho$_{67}$ with a half-life of 47 days in a storage ring. The conditions allowed obtaining a bare ^{163}Ds$_{66}^{66+}$- ion with 294 MeV/u, which decays into ^{163}Ho$_{67}^{66+}$- ion. Due to the only electron of Ho-ion, this ion could be quantitatively detected in a mixture with Ds-ion. Beta-bound decay has been experimentally demonstrated in the rhenium–osmium (^{187}Re–^{187}Os) system. Stable Re was transmuted to Os with a half-life of 33 years in a storage ring [4]. All of these examples do not have any practical application because their implementation requires a lot of energy. For example, in order to produce ions with kinetic energy of 294 MeV achieved in storage ring, it is necessary to heat the gas to 2.3E+12K. However, it is proven in these experiments that beta-decay rate varies under certain conditions.

3. Review of Experimental Studies on Detritiation of Tritium-contaminated Samples

The utilization of tritium-contaminated waste is very important, especially due to the fact that tritium is the fuel for ITER-International Thermonuclear Experimental Reactor, a new nuclear reactor that is being constructed in France by the global scientific and technical communities. Tritium decay with the generally accepted half-life of 12.32 years is based on the transmutation reaction resulting in formation of $_2$He^3, beta particle, and an antineutrino. This transmutation reaction can be monitored by measuring the amounts of He^3 formed and/or beta particles released. Most researchers have been measuring beta particles because it is much easier in many experimental conditions. However, measuring He^3 would make it easier to reach a conclusion about a rate of tritium decay.

Torikai et al. [5] studied the detritiation of tritium contaminated stainless steel samples using various purging gases (argon, air and argon +3 vol% H$_2$) and interpreted the results of the experiments assuming that a half-life of tritium decay has a generally accepted value. They purged samples heated up to a temperature of 673 K, collected tritium removed from these samples (hereafter, the removed tritium), determined the activity of the removed tritium, and stated that all sorbed tritium was removed from the sample. They determined an initial concentration of tritium in a sample indirectly based on the tritium content measured for another sample loaded at the same time, i.e., they did not know the exact amount of tritium sorbed in the sample studied, which made it very difficult to verify tritium balance after the test. They also did not measure the residual tritium activity of the sample after the completion of the detritiation, which made it impossible to verify the tritium balance during the test. However, they stated without any experimental proof that all sorbed tritium could be thermally released during heating the sample at a temperature higher than 673 K, which makes it difficult to accept their interpretation of tritium decay.

Akulov and Mamyrin [6] used mass-spectrometry for measuring He^3 concentration in gas phase during tritium decay experiments. They proved that the half-life for molecular tritium was by 11.5 days longer than the half-life for atomic tritium. Based on experimental data for atomic tritium decay and their model for possible atomic tritium decay reactions, they also calculated triton decay half-life that turned out to be by 9.5 days shorter than the half-life for atomic tritium decay. These data also prove that the experimental surrounding can affect beta decay rates.

Reifenschweller [7], in his experiments with tritium, counted beta particles formed during tritium decay and concluded that decelerated decay for beta radioactive tritium during the detritiation of a titanium sample (of 48 mg) consisting of mono-crystalline particles of about 15 nm in diameter arranged in chains and loaded with tritium in the hydrogen form (100 mCi T_2) was the only possible explanation for his experimental data. This study deserves a detailed discussion. The sample was slowly heated to 450°C within 10 h (at a specific rate per sample unit mass of 15.6°C/(min*g)), and the electron current was measured and attributed to beta particles released to gas phase of the volume where the required pressure was kept using a diffusion pump. It was shown that the electron current went down sharply to 72% of its initial value in a temperature range of 115–160°C followed by a slower decrease to 60% at 275°C. With a further temperature increase, the current returned to its initial value at 360°C, and then went down due to the complete decomposition of the titanium preparation, which indicated that most of tritium was removed from the metal sample. In another experiment, another sample was heated five times faster (at a specific rate per sample unit mass of 78.1°C/(min*g)) and there was no reduction in the electron current measured. The author stated that no tritium escaped from the titanium layer in the temperature range of 115–275°C, and this statement inevitably led him to conclude that the tritium decay constant decreases in this temperature range. The statement that "no tritium escaped from titanium layer in the temperature range of 115–275°C" was based on the fact that an increase in a temperature from 275°C to 360°C led to "re-increase of count rate." In order to prove this statement, it was needed to estimate tritium activity balance at each step of the process; however, the residual tritium inventory in titanium particles and a quantity of tritium released during the detritiation process and pumped out of the volume by the diffusion pump were not analyzed. Considering a small mass of titanium and its low tritium activity, it was probably impossible to measure in those experiments a concentration of tritium removed with the gas phase, a residual tritium activity of the sample, and/or a concentration of He^3 formed during tritium decay and remained in the sample after completion of the experiments. Finding tritium in the metal would be a proof that no tritium "disappearance" occurs, and a balance of the tritium activity in the experiments would allow one to estimate whether the tritium decay constant was changed. However, if the tritium balances for the experiments were not converged, and analytical procedures used for detection and measurement of tritium in all phases involved in the experiments were correct, it would be possible to conclude that beta bound decay or other nuclear reactions involving tritium occurred. However, there was no experimental proof there that the tritium decay constant was reduced.

Analysis of the aforementioned publications leads one to a conclusion that radioactive decay acceleration is possible dependent on the experimental surrounding and by applying an electrical potential to radioactive metals or accelerating the atoms to a very high speed. In contrast to previously suggested conclusions that the low temperature heating of metals either does not change a half-life of tritium [5], or even increases it [7], we demonstrate below that apparent tritium decay constant shows a completely opposite trend.

We are unaware about any study on transmutation of solid radioactive waste through nuclear reactions at low temperatures. However, the transmutation nuclear reaction could be one of the possible interpretations of the results of the research related to decontamination of tritium-contaminated metals [8]. We demonstrate below that it is possible to destroy radioactive isotopes by stimulating nuclear transmutation reactions.

4. Present Detritiation Experiments, Results and Discussion

The experiments described in details in [8] were conducted with stainless steel (dimension of 12.0 cm × 12.0 cm × 4.9 cm; mass of 5645 g), copper alloy Cu–Cr–Zr (dimension of 6.0 cm × 6.0 cm × 2.2 cm; mass of 710 g), tungsten (dimension of 4.0 cm × 4.0 cm × 1.0 cm; mass of 309 g), and beryllium (dimension of 3.0 cm × 3.0 cm × 1.0 cm; mass of 17 g). A brief summary of experimental setup is given below

The samples of the materials were cleaned using the same procedure as in the study [8] and loaded into the chamber, which was then pumped down and purged with argon to eliminate presence of residual air. The samples were heated
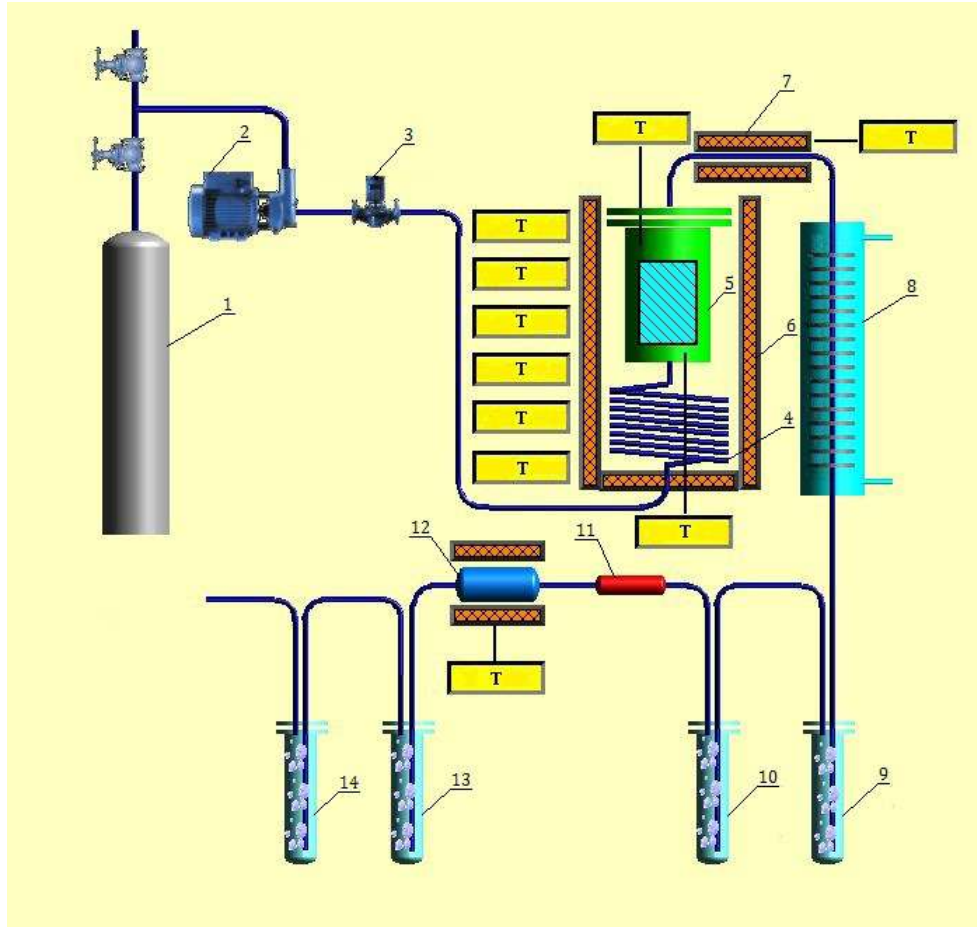
Figure 1. A block diagram of a facility for detritiation of metals: 1 – Cylinder with purging gas; 2 – Membrane pump/compressor; 3 – Gas flow meter; 4 – Heat exchanger; 5 – Chamber with a sample; 6 – Resistance oven; 7 – Heated communication lines; 8 – Condenser; 9, 10, 13, 14 – Bubblers; 11 – Dryer with absorbent; 12 – Catalytic reactor with heater.

for 10 h at a temperature of 773 K under a residual gas pressure of 10^{-4} P to remove gases dissolved in the metals. Samples were loaded with tritium by exposing them to a gaseous D–T mixture (48.3% of tritium, 50.6% of deuterium and 1.1% of protium) at a temperature of 473 K and a pressure around 0.05 MP for 24 h. The temperature was maintained uniformly through the whole chamber. The D–T gas mixture was rapidly evacuated from the chamber at the end of the exposure, and the chamber was immediately cooled down and purged with argon prior to opening.

Each detritiation experiment was conducted with one metal sample placed into a chamber where it was heated to a given temperature at a temperature increase rate of 10°C/min and simultaneously purged with a purge agent supplied to control a gas exchange rate (Fig. 1). The mixtures of 95 vol% argon + 5 vol% hydrogen, 95 vol% nitrogen + 5 vol% hydrogen, and argon + water vapor were used as purge agents. When a required operation temperature was achieved, the detritiation process continued for a duration ranging from 24 to 90 h, and the operation temperature was held stable during this period. The selection of a temperature increase rate of 10 ^{o}C/min was limited by the equipment capability.

Considering that (i) the mass of the smallest sample in our experiments exceeds by a factor of 350 the mass of each individual sample in Reifenschweller's experiments [7] and (ii) the operation temperature was steadily maintained in our experiments while was not steadily maintained in Reifenschweller's experiments, we concluded that a rate of the temperature increase in our experiments cannot affect the detritiation process because the duration of temperature increase period required to heat samples to the operation temperature contributed no more than 5.5% to the overall time of the detritiation process

The detritiation of each metal was studied at three different temperatures: 200°C, 500°C, and 800°C, with the three aforementioned purge agents supplied at various flow rates. A change in flow rate affects the kinetics of tritium desorption from metals and as such can affect other possible processes occurred inside the metal. During the detritiation experiments, the tritium transferred to gas phase was removed from the chamber with the flow of the purge agent and was collected in a tritium collection system consisting of a series of bubblers (9, 10, 13 and 14, Fig. 1) installed downstream of the chamber. The first pair of bubblers in a series was placed in between the chamber and the catalytic reactor filed with copper oxide. These bubblers trap tritium released in form of water vapor. The catalytic reactor operated at a temperature of about 770 K to oxidize gaseous hydrogen to water vapor. The second pair of bubblers was placed downstream to trap this water vapor that included tritium released by the samples in the form of gaseous hydrogen and converted in the catalytic reactor. The bubblers were designed to produce small bubbles of air and in order to ensure a high efficiency of the tritium collection.

The balance of tritium activity for each experiment was determined based on the following formula that represents material balance of that tritium in the experiment: A = B + C + D, where A is the initial tritium inventory in a metal; B is the residual tritium inventory in the metal after the experiment; C is the tritium inventory transferred to gas phase and collected during the experiment; and D is the possible tritium activity that could escape during the length of the experiment due to, for example, leakage through faulty air-tight barriers of an experimental system, and/or diffusion through the walls of the chamber. The radioactive decay of tritium is not considered here due to very short durations of the experiments when compared to the generally accepted half-life of 12.32 years.

Initial (A) and residual (B) tritium inventory and distribution in the samples were determined using radioluminography [9]. Tritium stripped from the metal in the detritiation experiments was very efficiently collected in the tritium collection system, and tritium content in both molecular hydrogen and water vapor forms was measured individually using the liquid scintillation counting method. The sum of tritium activity in its molecular hydrogen and water vapor forms (C) was calculated.

Before the experiments, the chamber with a metal sample loaded with tritium was checked with a helium leak detector and showed no leakage. During the experiments, the radiation level was measured in the premises surrounding the experimental system and was at a background level. After all experiments were completed, the walls of the chamber were cut, and the tritium activity and its distribution inside the walls of the reactor were measured. The result showed that tritium activity in the walls of the chamber was at a background level. These facts demonstrate that, within an accuracy of the measurements, the term D in the formula above is insignificant and was neglected in further analyses. Therefore, the difference between an initial inventory of tritium in a metal sample (A) and a sum of the residual inventory of tritium in the metal sample (B) and collected activity removed from the metal sample (C) was determined. This difference determined as percentage of A as $[1-(B+C)/A] \times 100\%$ is referred to hereafter as disbalance.

All metal samples studied had a tritium disbalance, which increased with an increase in temperature. Despite significant mass of the samples and tritium activity in them, the concentration of He ^{3} formed was so low that it was impossible to detect it. Tritium disbalances are dependent on gas exchange rates (the first number shown in parentheses is the optimal rate, after which a further increase does not improve tritium removal from the metals) at a temperature of 800°C as follows:

- Stainless steel – 47–95.2% (6.0–0.1 h $^{-1}$)

- Cu – 77–99.6% (6.0–0.1 h^{-1})
- W – 63-96.7% (6.0–0.1 h^{-1})
- Be – 87–95.3% (80–0.1 h^{-1})

Tritium disbalances at temperatures of 200°C and 500°C and optimal gas exchange rates determined at 800°C for each metal specified (the latter figures are shown in the parentheses) are:

- Stainless steel – 78.8% (200 °C, 6 h^{-1}) and 74.9% (500°C, 6 h^{-1})
- Cu – 45.8% (200°C, 6 h^{-1}) and 87.5% (500°C, 6 h^{-1})
- W – 39.1% (200°C, 6 h^{-1}) and 85.3% (500°C, 6 h^{-1})
- Be – 45.7% (200°C, 80 h^{-1}) and 94.6% (500°C, 80 h^{-1})

Attributing disbalance in the material balance equation shown above to the nuclear transmutation of tritium (via unspecified nuclear reactions and/or beta bound decay) and applying the exponential formula of the radioactive decay to these experimental results allows one to determine the half-life parameter describing the kinetics of tritium destruction achieved during the detritiation process ($T_{1/2cal}$) and compare it to the generally accepted half-life of tritium radioactive decay of 12.32 years. The ratios between the generally accepted half-life of tritium and the half-life parameter of tritium destruction obtained for various metals studied in these detritiation processes are in the following ranges:

- Stainless steel: 4.0E+3 – 2.0E+4 with the average ratio of 12.32/ $T_{1/2cal}$ = 1.0E+4
- Cu: 4.0E+3 – 3.5E+4 with the average ratio of 12.32/$T_{1/2cal}$ = 1.6E+4
- W: 3.2E+3 – 2.5E+4 with the average ratio of 12.32/$T_{1/2cal}$ = 1.5E+4
- Be: 3.9E+3 – 2.0E+4 with the average ratio of 12.32/$T_{1/2cal}$ = 1.5E+4

This means that the destruction of tritium as a radioactivity source via nuclear transmutation occurs in these experiments at a rate that is significantly faster than the generally accepted rate for the radioactive decay of tritium, i.e., the conditions of the experiments facilitate an acceleration of nuclear transmutation of tritium.

Exact mechanism of nuclear reactions involving tritium – accelerated radioactive decay and/or transmutation nuclear reactions with tritium participation – remains uncertain. However, regardless this mechanism, the observed phenomenon could be used for decontamination of tritium in tritium-contaminated metals in a controlled manner (partially recovering tritium for re-use and destroying the residual tritium).

Using the half-life parameters obtained for tritium destruction in stainless steel and considering a standard tritium decay mechanism only (with an average energy of β particles of 5.7 keV), energy released by 1 g of tritium per day will be very significant (4.4E+5 kJ/g), almost 10 000 times greater than for the same amount of practically any other fuel. As no experimental proof of this phenomenon is obtained, this conclusion should be considered as tentative. However, it hints for potential use of tritium energy released during destruction of tritium to self-sustain a process of tritium waste decontamination/destruction [10].

5. Conclusion

Tritium disbalances observed in these experiments can be explained by stimulated nuclear transmutation reactions and we are unable to offer another explanation of the experimental results. This phenomenon can be used for controlled decontamination of metals contaminated with tritium, and, tentatively, for self-sustaining a process of tritium waste decontamination/destruction by energy released there.

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