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(71) Applicant *(for all designated States except US)*: **THE STATE OF OREGON ACTING BY AND THROUGH THE STATE BOARD OF HIGHER EDUCATION ON BEHALF OF PORTLAND STATE UNIVERSITY** [US/US]; P.O. Box 751, Portland, OR 97207 (US).

(72) Inventor; and

(75) Inventor/Applicant *(for US only)*: **DASH, John** [US/US]; 313 N. Holman Street, Portland, OR 97217 (US).

(74) Agent: **SLATER, Stacey, C.**; Klarquist Sparkman, LLP, Suite 1600, One World Trade Center, 121 SW Salmon Street, Portland, OR 97204 (US).

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(54) Title: PROCESSING RADIOACTIVE MATERIALS WITH HYDROGEN ISOTOPE NUCLEI

(57) Abstract: A method for processing radioactive materials is disclosed. The method employs hydrogen isotope nuclei for the treatment of radioactive materials, such as uranium, and effectively increases the observed decay rate of such materials. Therefore, the disclosed method allows remediation of dangerous radioactive materials, such as uranium, without requiring long term, geologically-stable storage sites or costly, accelerator -based transmutation equipment.

PROCESSING RADIOACTIVE MATERIALS WITH HYDROGEN ISOTOPE NUCLEI

CROSS REFERENCE TO RELATED APPLICATION

This application claims the benefit of the earlier filing date of U.S. Provisional Application number 60/381,443, filed May 17, 2002, which is incorporated herein by reference.

FIELD

The disclosure concerns embodiments of a method for processing radioactive materials, with a particular embodiment comprising processing uranium with hydrogen isotope plasmas.

BACKGROUND

Disposal of spent nuclear fuel, transuranic elements and enriched uranium remain major impediments to the safe adoption of nuclear power sources. Many countries have accumulated significant amounts of hazardous radioactive waste from electricity and weapons production. Some components of this waste remain hazardous for thousands of years. By the end of 1998 over 38,000 metric tons of used or "spent" nuclear fuel had been accumulated in the United States from commercial power plants alone. According to Department of Energy (DOE) estimates, 2,500 metric tons of waste will be produced by the year 2035 from reactors under its supervision, including reactors for producing material for weapons, reactors for powering naval vessels, and research reactors. *See*, <http://www.em.doe.gov/idb96/tab13.html> (accessed May 16, 2003).

Currently, spent nuclear fuel and high level nuclear waste, such as partially purified nuclear fuel and surplus weapons grade materials, are stored at 129 different sites in 39 states. The different sites include commercial reactor facilities, research reactor sites, DOE/naval spent nuclear fuel and high-level radioactive waste sites, and surplus plutonium storage sites. Several of these storage sites are near cities, rivers, lakes, or seacoasts. Thus, these sites pose a threat to nearby populations and the environment.

There are two general proposed methods for remediating nuclear waste: (1) storage, and (2) transmutation. The DOE has proposed disposing of radioactive materials by long term geological storage. However, because the half life of elements contained in radioactive nuclear waste is so long, geological disposal requires a storage site or sites that will be geologically stable for thousands of years. For example, the half life of ^{235}U is 7.1×10^{8}

years; the half life of ^{238}U is 4.5 x 10^9 years; and the half life of ^{239}Pu is 24,000 years. Concerns about the effectiveness of waste confinement in geologic storage are overwhelming, and to date, no nation is using a permanent geologic site for storing high-level nuclear waste.

Transmutation has been proposed as an alternative to geological storage. Transmutation of one radionuclide into another can be accomplished by neutron bombardment in a nuclear reactor or in an accelerator-driven device. This typically proceeds by impinging a heavy metal target with a high-energy proton beam to produce a shower of neutrons by spallation. The neutrons can cause fission in a subcritical fuel assembly. But unlike a conventional reactor, fission ceases when the accelerator is turned off.

Some hazardous radioactive nuclides, such as plutonium 239 and the long-lived fission products technetium 99 and iodine 129, can be transmuted (fissioned, in the case of ^{239}Pu) with thermal (slow) neutrons. The minor actinides neptunium, americium and curium (as well as the higher isotopes of plutonium), which are all highly radiotoxic, are more readily destroyed by fissioning in the fast neutron energy spectrum. U.S. Patent Nos. 5,513,226 to Baxter et al., and 5,160,696, and 6,233,298, both to Bowman, provide examples of waste disposal by transmutation. Nevertheless, to date transmutation remains a theoretically attractive, but impracticable, method for disposing hazardous nuclear waste.

SUMMARY

The disclosure describes materials and embodiments of a method for processing radioactive materials for disposal. The method addresses the long-felt need for means to safely dispose of hazardous radioactive materials, such as materials used in weapons manufacture, nuclear reactors and research.

In one aspect, the method employs hydrogen isotope nuclei, such as hydrogen and deuterium nuclei, to treat a radioactive material. Any radioactive material can be treated according to the method, particularly materials that contain uranium isotopes and/or transuranic elements, such as plutonium. Specific working examples demonstrated the efficacy of the method for reducing the half life of uranium isotopes using hydrogen and deuterium nuclei.

In one embodiment, the method can be used to process spent nuclear fuel, which typically comprises additional, non-radioactive materials. Therefore, processing nuclear fuel may comprise separating uranium from the fuel, and exposing the uranium to a flux of hydrogen isotope nuclei. Separating the uranium from the fuel can comprise, for example,

dissolving the spent fuel in a molten salt to form a solution and electrorefining uranium and transition metals from the solution.

In one embodiment, the method employs hydrogen isotope nuclei obtained by glow discharge of hydrogen gas, deuterium gas, or both. In another embodiment the method uses electrolysis to produce hydrogen isotope nuclei from water. In one aspect, the water is deuterated or enriched in deuterium, and therefore electrolysis of the water yields deuterium nuclei in addition to hydrogen nuclei (protons).

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagram of an apparatus used for glow discharge mediated production of hydrogen isotope nuclei and treatment of radioactive materials with the nuclei.

FIG. 2 is a diagram of an electrolytic cell used to treat samples with protons or deuterons.

FIG. 3 is a bar graph illustrating the intensity of alpha radiation of uranium samples following exposure to hydrogen isotope plasmas versus alpha radiation emitted by a control sample.

FIG. 4 is a bar graph illustrating the intensity of beta particle radiation from four hydrogen isotope plasma-treated samples versus a control sample, measured at four different times.

FIG. 5 is a bar graph illustrating the intensity of gamma radiation from four hydrogen isotope plasma-treated samples versus a control sample, measured at three different times.

FIG. 6 is a bar graph illustrating the intensities of characteristic thorium 231 and thorium 234 gamma ray emissions from hydrogen isotope plasma-treated samples.

FIG. 7 is a bar graph illustrating the intensities of characteristic X-ray emissions from hydrogen isotope plasma-treated samples.

FIG. 8 is a bar graph illustrating the intensities of ^{235}U characteristic gamma radiation from hydrogen isotope plasma-treated samples.

FIG. 9 is a scanning electron microscope (SEM) micrograph of a uranium sample after exposure to deuterium plasma.

DETAILED DESCRIPTION

This disclosure includes data demonstrating that the decay rate of radioactive elements can be increased. Specifically, treatment of radioactive materials with hydrogen isotope nuclei, such as those produced in glow discharge and electrolysis of water, results in

accelerated decay of the radioactive materials. In a working embodiment, the observed decay rate of uranium was more than doubled by treatment with deuterium plasma.

I. Processing Radioactive Materials

According to the method disclosed herein, radioactive materials are processed for disposal so that the time required for the materials to decay to a safe level is reduced. Exposure of radioactive materials to a flux of hydrogen isotope nuclei induces accelerated decay of the materials. Materials exposed to the hydrogen isotope nuclei for a few hours exhibit accelerated decay. Indeed, exposure times suitable for accelerating the decay of radioactive materials range from less than one hour to hundreds of hours. In working embodiments radioactive materials were exposed to hydrogen isotope plasmas for periods ranging from about 18 hours to about 550 hours.

II. Sources of Hydrogen Isotope Nuclei

Hydrogen isotope nuclei, such as nuclei of hydrogen and deuterium, suitable for accelerating the decay of radioactive materials can be produced by several methods, and all such methods are suitable for generating nuclei for treating radioactive materials as described in the present disclosure. Specific examples of nuclei sources include plasmas and electrolysis processes. Exemplary plasma sources suitable for preparing hydrogen isotope plasmas are taught by U.S. Patent Nos. 6,441,553 to Yializis et al., 6,433,480 to Stark et al., and 6,059,935 to Spence et al., which are incorporated herein by reference.

The particular glow discharge apparatus used in working embodiments is depicted in Fig. 1. With reference to Fig. 1, conduits 1 and 2 are provided for optional coolant flow; input power is provided at to anode 4 at input 3; conduit 5 provides optional coolant flow for cooling quartz tube 9; sample (not shown) is placed between anode 4 and cathode 8 and secured to the face of cathode 8 via a molybdenum nut (not shown).

In one embodiment the source of hydrogen isotope nuclei can be an electrolysis apparatus. For the production of protons, H_2O can be subjected to electrolysis conditions, and the radioactive material can be used as an electrode for the electrolysis reaction so that the material is subjected to *in situ* proton treatment. Similarly, for treatment with deuterons, D_2O, or deuterium enriched water can be electrolyzed. The electrolysis of water does not require high power or any complex or specialized apparatus. For example, working embodiments employed a current of 1.5 amperes at a potential of about 4 volts and used the electrolysis cell depicted in Fig. 2. With reference to the electrolysis cell of Fig. 2, container 11 contains water 12 with dissolved electrolyte, such as a strong acid, in working

embodiments sulfuric acid was used as the electrolyte; anode 13 is attached to an external power source (not shown); recombination catalyst 14 catalyzes the reaction of excess hydrogen (or deuterium) with oxygen to give water; teflon cap 15; cathode lead wire 16 (also connected to an external power source) is connected to uranium cathode 18 via platinum foil welded to lead wire 16; and line 19 indicates the electrolyte level.

III. Radioactive Materials

In principle the decay of any radioactive material can be accelerated by treatment according to the method disclosed herein. Currently, the bulk of hazardous radioactive waste comprises uranium and/or transuranic elements, and the present method is well suited for processing such materials. Generally, the term "transuranic" refers to any radioactive element having an atomic number greater than that of uranium (92). One example of a transuranic element is plutonium.

Additional examples of radiation emitting isotopes that can be processed by the disclosed method include isotopes of other elements, such as phosphorus, sulfur, hydrogen, carbon, indium and iodine. Particular examples of radiation emitting isotopes include, but are not limited to, phosphorus 32, sulfur 35, tritium (^{3}H), carbon 14, indium 111, iodine 121, uranium 235, uranium 238 and plutonium 239. In particular examples of the disclosed method, the decay of uranium, particularly uranium 238 was enhanced.

IV. Results

Uranium metal treated by the method disclosed herein exhibits increased flux of alpha, beta and gamma radiation, and consequently exhibits an increased effective rate of decay. With respect to Fig. 3, the rate of alpha particle emission per gram of sample indicates that exposure of uranium to hydrogen isotope nuclei, particularly hydrogen isotope plasmas, greatly increases the alpha radiation flux. The flux from three of the samples is almost the same and is more than twice the flux from the control. Two of these samples were exposed to hydrogen plasma and one was exposed deuterium plasma. The flux from the fourth sample, which was exposed to deuterium plasma for ~ 550 hours, is even greater, although not as great as shown in Fig. 3, due to spalling of the sample surface. That is, the surface areas of the control sample and samples H1, H2, and D1 are proportional to mass, but this is not true for sample D2 because of the spalling which greatly reduced the mass and increased the surface area. Any increase in normalized alpha particle flux is useful, for example, from about 5% to greater than 500%, working embodiments resulted in an increase in alpha particle flux of from greater than 100% without changing the surface area

of the sample. Because the increased flux indicates an increased rate of decay, the present method has applicability to the disposal of radioactive materials. Moreover, the increased flux observed indicates that the material treated with hydrogen isotope nuclei can serve as nuclear fuel with less, or even without, enrichment being required. Thus, nuclear fuel can be prepared from non-enriched uranium by processing the uranium according to embodiments of the present method.

Fig. 4 shows the results of beta radiation measurements recorded periodically over a span of about one year. The beta emissions did not change with time for any of the samples during this period. All of the samples exposed to hydrogen isotope plasmas had larger weight-normalized beta fluxes than the control. For H1 and H2, the increases were 9.6% and 7.7%, respectively. For D1 and D2, the increases were 15.4% and 45.4%, respectively. As a result, working embodiments indicate that beta flux increased for working embodiments by at least about 5% and as much as about 50% relative to an untreated control.

All of the samples exposed to hydrogen isotope plasmas also exhibit higher flux of high energy gamma and X-rays than the control. With reference to Fig. 5, the observed increases in gamma ray emission per gram are about 10% for sample H1, 5% for H2, 14% for D1, and 59% for D2. Greater percentage increases are possible, such as a greater than 100% increase in gamma flux per gram. In working embodiments useful increases in gamma radiation flux were observed from about 5% to about 60%.

Eleven of the most prominent characteristic spectral peaks observed with the gamma ray spectrometer were analyzed to determine correlations with the alpha, beta and gamma emissions. The intensities of each of the eleven peaks for the four plasma-treated samples and the control are recorded in Figs. 6–8. With reference to Fig. 6, the first peak in the gamma ray spectrum, at 63.1 keV, was from thorium 234, which results from alpha decay of uranium 238. The second peak, at 84.1 keV, was from thorium 231, which results from alpha decay of uranium 235. The third peak, at 92.4 keV, is also from the decay of thorium 234. The observed intensity for every peak from the reacted samples is greater than for the corresponding peak from the control, which indicates an increase in the decay product for treated samples relative to the control. The increased intensity is greatest for the 92.4 keV peaks, which indicates an increase in the amount of thorium 234. The intensity of this peak for H1 was 18% greater than the control, for H2 it was 19% greater, for D1 it was 33% greater, and for D2 it was 39% greater. Any increase in the amount of thorium 234 is advantageous, thus increases of from about 1% indicate a desirable increased decay of uranium 238. In working embodiments the increased decay results in increased amounts of

thorium 234 ranging from greater than about 10% to at least 40% greater relative to an untreated control sample.

With reference to Fig. 7, the next four most intense peaks (after those analyzed in Fig. 6) were identified as characteristic X-ray peaks from uranium, namely, Kα2 at 94.9 keV, Kα1 at 98.5 keV, Kβ1 at 111.1 keV, and Kβ2 at 114.8 keV. Fig. 7 illustrates that all of the Kα2 and Kβ4 peaks for the reacted samples are **less** intense than the corresponding peaks for the control. Therefore, this data establishes that the samples contain less uranium per unit mass than the control. The average reduction in intensity is 13% for H1, 7% for H2, 24% for D1, and 37% for D2. In the case of Kα1, the reduction is 4% for H1, but H2 has about 1% greater intensity than the corresponding control peak. Sample H2 was exposed to glow discharge for only 18 hours, far less than the other samples. Both D1 and D2 have reduced Kα1 intensities, 8% and 28%, respectively, compared with the control. For the Kβ1 peak, the intensity of H1 is the same as the control, H2 is 7% higher, D1 is 4% higher, but D2 is 26% lower. The large decreases in observed intensity for characteristic uranium X-ray peaks for the plasma treated samples H1, H2, D1 and D2 indicate the method results in reduced concentration of uranium in the samples.

At higher energy in the gamma ray spectra, the next four prominent peaks are gamma rays emitted by uranium 235. The intensities of these peaks for the four reacted samples and for the control are recorded in Fig. 8. The differences between the treated samples and the control are not as great as those observed for the thorium 231 and 234 characteristic gamma ray peaks. Nonetheless, here, as before, the gamma ray emissions are greater in every case for the reacted samples H1, H2, D1, and D2 than for the control. By far the largest increases in emission were from sample D2 which was subjected to the glow discharge plasma much longer than the other samples.

The plasma-exposed samples and the control were analyzed using thermal ionization mass spectrometry (TIMS) at the Massachusetts Institute of Technology Radiogenic Isotope Geochemistry Laboratory, using a VG Sector 54 multicollector mass spectrometer, to determine changes in isotopic composition induced by treatment of the uranium samples with hydrogen isotope nuclei. The results are recorded in Table 1, below. With reference to Table 1, the ^{206}Pb / ^{204}Pb ratio for each of the plasma-exposed samples exceeds this ratio for the control by more than three standard deviations. Lead 206 is a stable isotope formed at the end of the uranium 238 decay chain, and uranium 238 is the most abundant uranium isotope, comprising about 99.3% of natural uranium. Uranium 238 decay proceeds through 13 different unstable isotopes and terminates with the stable isotope, lead 206. However, lead 204 is not a decay product and therefore the amount of

this stable isotope is constant. Thus, the ^{206}Pb / ^{204}Pb ratios recorded in Table 1 indicate that uranium 238 and its daughter products are decaying more rapidly in each of the four plasma-exposed samples than in the control.

With reference to the ^{206}Pb / ^{204}Pb ratios recorded in Table 1, indicate an increase in the ratio of from about 0.7% to about 1.2%. These measurements were performed approximately 18 months after treatment of the samples with hydrogen isotope plasmas. Of course, this ratio will increase as the remaining uranium 238 in the sample decays, and calculations indicate that about half of the uranium will have decayed after about 100 years.

TABLE 1

Pb Isotopic Ratios

Sample	206 /204	σ	207 / 204	σ	208 /204	σ	208 /206	σ
Control	18.061	0.025	15.578	0.028	37.591	0.027	2.0813	0.012
H1	18.193	0.026	15.554	0.028	37.766	0.029	2.0759	0.010
H2	18.493	0.079	15.607	0.070	38.052	0.071	2.0577	0.025
D1	18.534	0.102	15.628	0.097	38.154	0.122	2.0586	0.091
D2	18.286	0.031	15.587	0.031	37.757	0.032	2.0648	0.017

With reference to Table 1, there is no significant change in the ^{207}Pb / ^{204}Pb ratio for the plasma-exposed samples compared with the control, probably because lead 207 results from decay of uranium 235, which has an abundance of only 0.7%. Any change in the ^{207}Pb / ^{204}Pb ratio likely would be beyond the detection limit of the spectrometer used. There is a significant increase in the ^{208}Pb / ^{204}Pb ratio for each of the plasma-exposed samples. Because lead 208 is not in the decay chain of uranium, the increase must be from another, source. The data in Table 5 also show that the ^{208}Pb / ^{206}Pb ratio is slightly less for each of the plasma-exposed samples than for the control, even though the natural abundance of lead 208 is more than twice that of lead 206. Thus, the concentration of lead 206, the decay product of uranium 238, is produced at a higher rate in the plasma-treated samples. This indicates that uranium 238 radioactive decay rate is increased in the plasma-exposed samples relative to the decay rate in the untreated sample. Thus, the empirical results clearly establish that the effective decay rate of uranium 238 and uranium 235 is significantly increased by processing with hydrogen isotope nuclei according to

embodiments of the present method. The alpha, beta, and gamma radiation data and the mass spectrometer data presented above indicate that the interaction of uranium with hydrogen isotope nuclei during glow discharge and electrolysis increases the rate of radioactive decay which results in shortening of the decay time of uranium.

Figs. 3 and 4 contain radiation measurements made over a period of six months. These data show that the accelerated rates of decay of the uranium samples treated with hydrogen isotope nuclei has not diminished over this time period. Thus, the effective half life for uranium decay in these samples will be less than that of natural uranium. The change in half life can be estimated from the approximately 1% reduction in uranium content of the samples after exposure to glow discharge. The TIMS analyses were performed about 18 months after the glow discharge exposures. According to TIMS analyses, there was an approximately 1% reduction in uranium content over 18 months. From this reduction, a simple calculation suggests that the half life of uranium 238 was reduced from 4.5×10^9 years for uranium not processed according to disclosed embodiments to about 100 years for processed uranium.

V. Discussion

A different interpretation of the TIMS mass spectrometer data could be suggested. For example, the lower uranium content of the plasma-exposed samples could be caused by greater oxygen content (possibly due to the presence of uranium trioxide and/or triuranium octaoxide), due to greater surface areas. The basis for this interpretation is that each of the plasma-exposed samples has greater surface area than the control, due to roughening caused by sputtering. The surface area is proportional to the plasma exposure time, because current density was approximately constant and the same for all samples. However, if this explanation were correct, then sample D2 would have by far the greatest oxygen content and the lowest uranium content because about half of its mass was lost due to sputtering, necessitating the use of two, three mm diameter discs for the TIMS analysis in order to have mass similar to the other samples. This means that D2 had more than twice as much surface area as each of the other samples. Therefore, the oxygen content of D2 was potentially at least twice that of the other samples, and the uranium content of D2 should have been reduced at least twice as much as the other plasma-exposed samples. In fact, Table 5 shows that all of the plasma-exposed samples have about 1% less uranium than the control, and this data is consistent with the reduced characteristic X-ray intensities of the plasma-exposed samples given in Fig. 7. Thus, the alternative interpretation forwarded above does not explain the results presented herein.

EXAMPLE 1

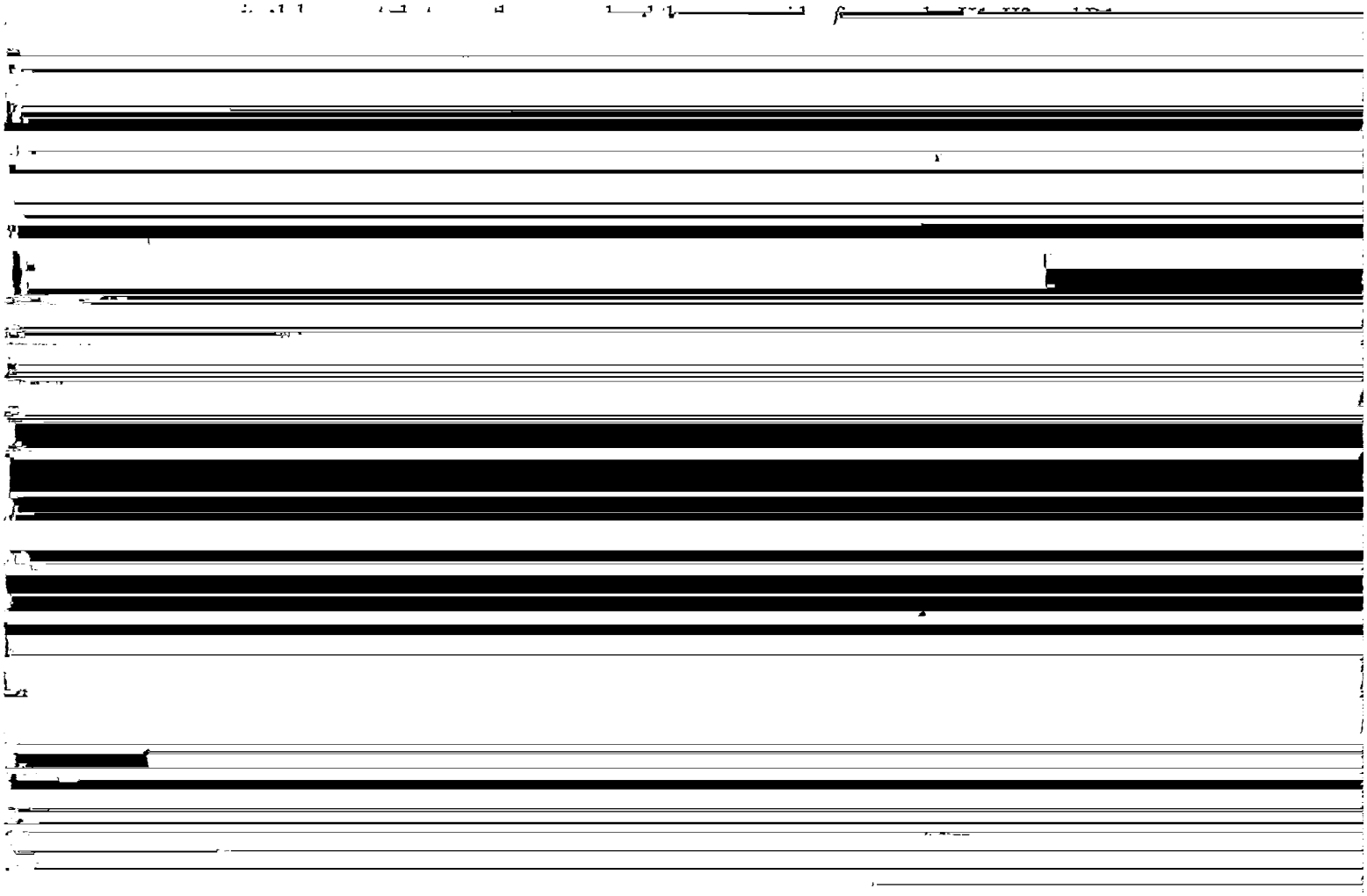
This example describes treating uranium metal with hydrogen isotope plasmas. A uranium disk (99.98% purity) measuring 19 mm (diameter) by 0.2 mm (thickness) was placed in the glow discharge apparatus diagrammed in Fig. 1. The uranium disk was held against the face of the cathode with a molybdenum nut. The nut shielded 53% of the uranium disk's face area (the diameter of the exposed area was 13 mm). Glow discharge was performed at a gas flow of 5 torr, using a 5 milliampere current at a potential of 500 volts. Four samples were treated to glow discharge using either a hydrogen or a deuterium plasma. Sample H1 was treated with hydrogen plasma for 43 hours; sample H2 was treated with hydrogen plasma for 18 hours. Samples D1 and D2 were treated with deuterium plasma for 100 and 550 hours, respectively.

EXAMPLE 2

This example describes the characterization of the samples treated with hydrogen isotope plasmas according to Example 1. Alpha, beta, and gamma measurements were performed on each sample following treatment with the plasma, and the results were compared to those obtained from an untreated uranium foil as a control. The alpha particle measurements were performed using a Ludlum 43-5 alpha probe, and the beta measurements were made using a Ludlum model 2000 GM counter (both instruments are available from Ludlum Instruments, Sweetwater Texas). Gamma and X-ray spectra were obtained using an EG&G Ortec 92X gamma ray spectrometer (available from Ortec, http://www.ortec-online.com/index.html). Alpha, beta, gamma and X-ray measurements were taken of each sample over approximately one year. The results were normalized to the weight for each sample, and are recorded in Figs. 2–4.

EXAMPLE 3

This example describes the characterization of alpha particle emission from the control and plasma-treated samples of Example 1. The alpha radiation emitted in one hour by uranium foil control sample and by samples exposed to hydrogen isotope plasmas during glow discharge is recorded in Table 2, below. The original uranium sample disks were cut from a sheet with a steel punch. This produced a convex surface on one side and a concave surface on the reverse side, which resulted in a shorter path length from the convex surface to the detector, and about a 20% increase in alpha counts compared with the concave surface. The samples used in the glow discharge apparatus were flattened during attachment to the cathode. With reference to the data recorded in Table 2 there is almost no difference



despite the fact that the exposed surfaces of these samples were roughened by sputtering during glow discharge. Sample D2, which was in the glow discharge plasma much longer than the other samples, has about a 7 % higher net count of alpha from the exposed compared with the reverse side. One explanation for this observation is that the exposed surface is much rougher, which would enhance the emission of alpha radiation due to the increased surface area. Sample D2 has the highest net counts of alpha particles observed among the five samples. Thus, Table 2 establishes that the observed increase in alpha radiation for uranium samples processed according to the embodiments of the disclosed method is not due to surface roughening.

TABLE 2

Sample	Mass (g)	Upside	Net Counts
Control	0.9904	convex	13729
		concave	11182
H1(43h)	0.9293	exposed	26960
		reverse	28095
H2(18h)	0.7645	exposed	21506
		reverse	21290

the presence of 1% thorium with a standard deviation of 0.3%. The presence of 1% thorium in the sample indicates that uranium 238 is decomposing at a rate effective to produce the thorium.

EXAMPLE 5

This example describes mass spectrometric analyses of the plasma-exposed samples and the control to determine if isotopic and/or elemental content of the samples was changed by the plasma treatment. 25 mg portions of the plasma-exposed samples and of the control, were analyzed using an Elemental Axiom SC magnetic sector inductively coupled plasma mass spectrometer (ICPMS) at Oregon State University, in the W.M. Keck Collaboratory for Plasma Spectrometry. Three analyses were performed on the control and on each of the samples exposed to hydrogen isotope plasma. Lead was detected at a level of about 100 ppm in each of the plasma-exposed samples but not in the control. The results are given in below in Table 3. Average counts for Pb and standard deviations (σ) for three ICPMS analyses for a blank and for each of five uranium samples are provided. The results recorded in Table 3 indicate that the blank and the control uranium sample have almost the same number of counts for each of the three lead isotopes. However, the four samples subjected to plasma exposure all had significant increases in the average number of counts for all three lead isotopes.

TABLE 3

Sample	**Pb 206**		**Pb 207**		**Pb 208**	
	Average	σ	Average	σ	Average	σ
Blank	36816	2275	31451	1279	78224	1862
Control	36118	5876	30120	5052	73491	12022
H1(43h)	48684	13850	40900	9834	102889	22699
H2(18h)	52304	11158	43253	9388	107882	22091
D1(100h)	51211	16997	45664	9116	117802	12491
D2(550h)	45773	9338	39171	8463	96442	20577

The ratio of Mass234/Th232 also was determined for each sample using ICPMS. This ratio was much larger for the four plasma-exposed uranium samples than for the control, as shown below in Table 4, which also includes the standard deviations (σ) for the recorded Mass234/Th232 ratio analyses.

TABLE 4

Sample	Analyses	Av. Mass234/Th232 Ratio	σ
Control	3	41.4	2.3
H1(43h)	3	61.0	4.0
H2(18h)	3	55.7	5.3
D1(100h)	3	61.0	5.4
D2(550h)	3	55.3	7.6

All of the 12 analyses on the plasma-exposed samples have higher Mass234/Th232 ratios, and the recorded ratios exceed 3σ of the control for 11 of the 12 analyses. The twelfth analysis exceeds 2σ of the control. These higher ratios appear to have been caused by depletion of Th232 in the plasma-exposed samples, while the mass 234 content increased. For example, there was an increase of about 3% in mass 234 content and about 16% decrease in Th232 content of sample H2(18h) compared with the control. Hydrogen and deuterium plasmas appear to be equally effective in increasing the mass234 / ^{232}Th ratio. For the times involved in these experiments, there is no correlation between the magnitude of this ratio and the time of exposure to the plasma. Mass 234 is a combination of thorium 234, protactinium 234, and uranium 234, all of which are in the decay chain of uranium 238. These isotopes cannot be distinguished by ICPMS. The increase in the mass234 / ^{232}Th ratio relative to the control indicates that uranium 238 decay is accelerated.

EXAMPLE 6

This example describes the determination of uranium content, and the ratios of lead isotopes in each plasma-treated sample and the control sample by TIMS. Analyses were performed at the Massachusetts Institute of Technology Radiogenic Isotope Geochemistry Laboratory, using a VG Sector 54 multicollector mass spectrometer. Discs three mm in diameter were punched from each sample and sent to this laboratory. Highly eroded sample D2(550h) was only about one-half the thickness of the others, so it was necessary to provide two three mm diameter discs. The analyses were performed blind: thus, the analysts had no knowledge of the prior history of each sample. The only nuclides detected in the mass range 230-240 amu were uranium 234, uranium 235, uranium 236, and uranium 238. All samples were found to have the same isotopic composition, within the analytical uncertainty, and this composition was the same as that of natural uranium. Thorium 232 was not detected in these analyses because the procedure for purification of uranium prior to uranium isotope analysis separates thorium from uranium, so no thorium remains in the analytical sample.

The total uranium in each sample, as determined by TIMS, is given in Table 5, below, which establishes that the control sample contains about 1% more uranium than the samples exposed to hydrogen isotope plasmas. These results are consistent with the data given in Fig. 7, where 13 of 16 characteristic X-ray peaks from the plasma-exposed samples have lower intensities than the corresponding peaks from the control sample. Sample D2 has the lowest intensities for all four characteristic X-ray peaks and it also has the lowest uranium content.

TABLE 5

Sample	Mass (mg)	U in sample (mg)	% U in Sample
Control	24.50	24.45	99.79
H1(43h)	27.05	26.65	98.51
H1(43h)R	27.05	26.71	98.75
H2(18h)	26.60	26.30	98.86
H2(18h)R	26.60	26.22	98.56
D1(100h)	21.60	21.34	98.80
D2(550h)	25.75	25.34	98.41

With respect to Table 5, H1(43h)R and H2(18h)R are repeat analyses, which were performed in order to verify the precision of the results. The average U content of treated samples H1, H2, D1 and D2 is 98.6 +/- 0.3 % U, whereas the control contains 99.8+/- 0.3 % U. Therefore, the difference between these averages is about 1.2%. Even considering the maximum error, the difference is statistically significant.

The data obtained from TIMS analyses performed upon each plasma-treated sample and the control for lead isotopes are given in Table 1 in terms of the ratio of the amount of each of the three most abundant isotopes to the amount of lead 204, a stable isotope which is not produced by radioactive decay of uranium. Table 1 also lists the observed ^{208}Pb / ^{206}Pb ratios. This data establishes that the production of lead isotopes by decay of uranium isotopes is increased in the plasma-exposed samples relative to the control.

EXAMPLE 7

This example describes using electrolysis of water to provide a source of hydrogen isotope nuclei. With reference to Fig. 2, cathode 8 was natural (99.98% pure) uranium foil, 20 mm long, 4 mm wide, 0.18 mm thick. The anode 3 was a circular disk of high purity, thin platinum foil, having a 2.6 cm diameter. The electrolyte 2 contained about 3 ml reagent grade H_2SO_4 in about 60 ml of high purity (99.9%) D_2O. Cell 1 was a 200 ml Pyrex beaker;

Pt anode 3 was attached to a 1 mm diameter Pt wire by spot welding, after which the weld was cleaned by immersion in H_2SO_4. A 1 mm diameter Pt lead wire also was used for the cathode 6. A Pt foil 7 was spot welded to the lead wire, and the weld was cleaned by immersion in H_2SO_4. Uranium foil 8 was bent over the Pt foil and crimped. In this way it was possible to avoid contamination, which might have resulted from vaporization of uranium during spot welding. The Pt wires were threaded through the tapered Teflon cell top 5 for connection to the electrical circuit. A diagram of the electrolysis cell is shown in Fig. 2.

Electrolysis was performed for about 5 hours at a constant current of 1.5 amperes and a cell voltage of about 4 volts. The uranium foil was then removed from the cell, rinsed in deionized water, and air dried. Radiation emissions from the electrolyzed sample and from an unelectrolyzed control sample cut from the same 20 x 25mm foil were measured with a Ludlum Model 3 survey meter. The meter is equipped with a model 44-9 probe (a pancake detector with a thin mica window), which can detect alpha, beta, and gamma radiation. The electrolyzed uranium was found to emit a significantly higher flux of radiation than the control. These measurements were made periodically over the next eight months, and the results were the same. There was no detectable change in the flux of radiation from either sample.

The samples also were analyzed at the Reed College Reactor. Measurements were made blind, i.e., the scientists who performed the measurements had no knowledge of the prior history of the samples. The alpha measurements were made with a Ludlum 43-5 alpha probe. The beta plus gamma measurements were made with a Ludlum model 2000 GM counter. By placing a 1 mm thick aluminum plate between the detector and the sample, it was possible to detect the continuum of high energy gamma and X-rays. The results are provided by Tables 6 to 8, where the uranium cathode is labeled JD1-after.

Table 6 includes beta-plus-gamma radiation data obtained by Reed Reactor scientists from an unelectrolyzed control and from two electrolyzed uranium samples, both of which have significantly greater specific radiation emission (counts per second per gram of uranium, c/s·g). The data provided by Table 6 establishes that at least a 10% increase and up to at least a 15% increase in emitted radiation occurs with samples treated according to the hydrogen isotope nuclei produced by electrolysis.

TABLE 6

Sample	Sample mass, g	Counts Beta+Gamma	Sigma B+G	B+G specific c/s·g	% increase
Control	0.3104	169233	411	1816	
JD1-after	0.2904	175581	419	2014	11
DC3-after	0.1543	96490	311	2082	15

A second electrolysis experiment was performed where, in order to minimize corrosion at the contact between the uranium cathode and the platinum lead wire, the circular anode was placed on the bottom of the cell, and the contact was covered with Dow Corning 732 silicone rubber sealant. This prevented the electrolyte from contacting the upper part of the uranium cathode.

The electrolyte for this experiment contained 3 mL concentrated H_2SO_4 and 60 ml of deionized H_2O. The current was 35 milliamperes and the current density was about 26 milliamperes/cm^2. The cell voltage was about 2.4 volts and the time of electrolysis was about 65 hours. After electrolysis, the uranium cathode was rinsed in deionized water and dried, and then the silicone sealant was removed. The uranium was then cut at the interface between the electrolyzed and the unelectrolyzed parts. Then radiation measurements were made by Reed Reactor scientists. The unelectrolyzed part showed no increase in any type of radiation, but the electrolyzed part had significant increases in specific radiation. The results are given in Tables 6–8 for sample DC3-after.

TABLE 7

Sample	Sample mass, g	Gamma counts	Sigma gamma	Gamma, specific, c/s·g	% increase
Control	0.3104	21055	145	225	
JD1-after	0.2904	21717	147	248	10
DC3-after	0.1543	11519	107	246	9

Table 7, includes gamma radiation data obtained by Reed Reactor scientists from an unelectrolyzed control and from two electrolyzed uranium samples, both of which have significantly greater emission rates than the control. This establishes that an increase of 10% in gamma radiation emission can be induced by treatment with hydrogen isotope nuclei produced by electrolysis.

TABLE 8

Sample	Sample mass, g	Alpha counts	Sigma counts	Alpha specific, c/s·g	% increase
Control	0.3104	4104	64	44	
JD1-after	0.2904	5057	71	58	32
DC3-after	0.1543	2657	52	57	31

Table 8 includes alpha radiation measurements obtained by Reed Reactor scientists from an unelectrolyzed control and from two electrolyzed uranium samples, both of which have significantly greater emission rates than the control.

The present application has been described with reference to several embodiments. These embodiments are considered exemplary only, and the scope of the invention should not be considered limited to these embodiments.

I claim:

1. A method for processing a radioactive material, comprising:
providing a source of hydrogen isotope nuclei; and
contacting the radioactive material with a flux of the hydrogen isotope nuclei.

2. The method according to claim 1 where the decay rate of the radioactive material is increased.

3. The method according to claim 1 where the source of hydrogen isotope nuclei is a plasma.

4. The method according to claim 1 where the source of hydrogen isotope nuclei is electrolysis.

5. The method according to claim 1 where the radioactive material comprises uranium.

6. The method according to claim 5 where the radioactive material comprises an increased ratio of lead 206 to lead 204 after processing.

7. The method according to claim 5 where the radioactive material exhibits increased flux of at least one emission selected from the group consisting of alpha particles, beta particles and gamma rays after processing.

8. The method according to claim 7 where the material exhibits increased gamma ray flux of from about 5% to about 60% after processing.

9. The method according to claim 7 where the material emits from about 5% to about 50% more beta radiation per gram of the material after processing relative to the material prior to processing.

10. The method according to claim 9 where the material emits from about 15% to about 45% more beta radiation per gram of the material after processing relative to the material prior to processing.

11. The method according to claim 7 where the material emits from about 5% to about 100% more alpha radiation per gram of the material after processing relative to the material prior to processing.

12. A method for processing spent fuel from a nuclear reactor, comprising:
providing spent fuel comprising uranium; and
exposing the uranium to a flux of hydrogen isotope nuclei, whereby the decay rate of the uranium is increased.

13. The method according to claim 12 where the flux of hydrogen isotope nuclei is produced by glow discharge of hydrogen gas, deuterium gas, or both.

14. The method according to claim 12 where the flux of hydrogen isotope nuclei is produced by electrolysis of water, deuterium enriched water, or both.

15. A method for accelerating decay of a radioactive element, comprising:
providing a proton source; and
contacting the radioactive element with protons from the proton source.

16. The method according to claim 15 where the proton source is a hydrogen plasma.

17. The method according to claim 15 where the protons are produced by electrolysis of water.

18. The method according to claim 15 where the radioactive element is uranium.

19. A non-enriched nuclear fuel, comprising processed uranium having an increased flux of at least one emission selected from the group consisting of alpha particles, beta particles and gamma rays relative unprocessed, non-enriched uranium.

20. The non-enriched nuclear fuel according to claim 19 where the processed uranium emits from about 5% to about 50% more beta radiation per gram relative to the uranium prior to processing.

21. The non-enriched nuclear fuel according to claim 19 where the processed uranium emits from about 5% to about 100% more alpha radiation per gram of the uranium after processing relative to the uranium prior to processing.

5

22. The non-enriched nuclear fuel according to claim 19 where the processed uranium emits from about 5% to about 60% more gamma radiation per gram of uranium relative to the uranium prior to processing.

FIG. 1

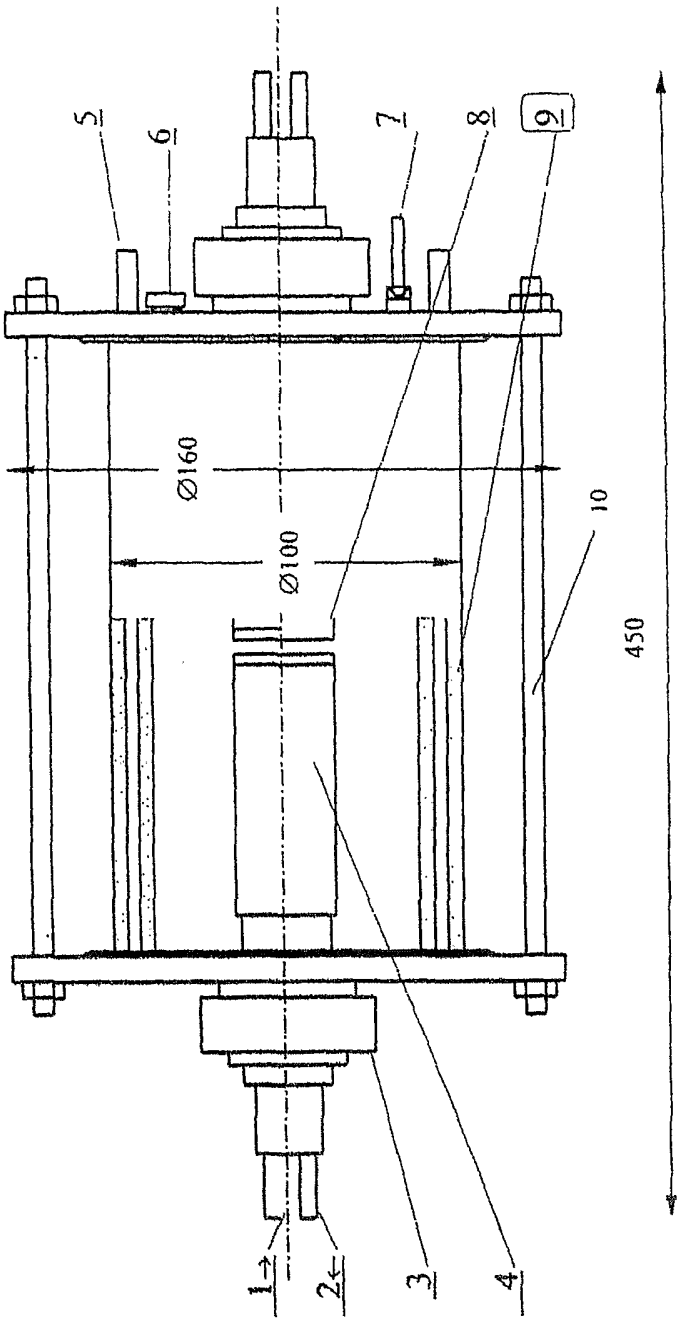


FIG. 2

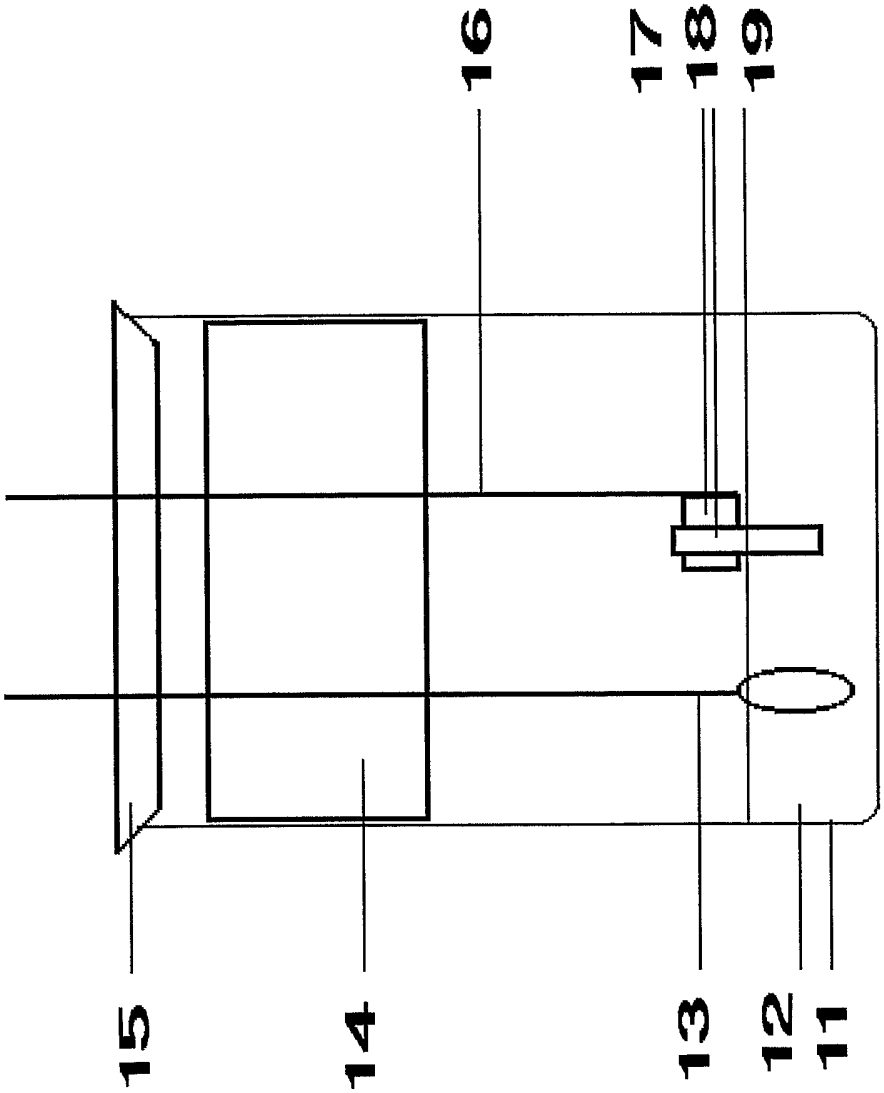


FIG. 3

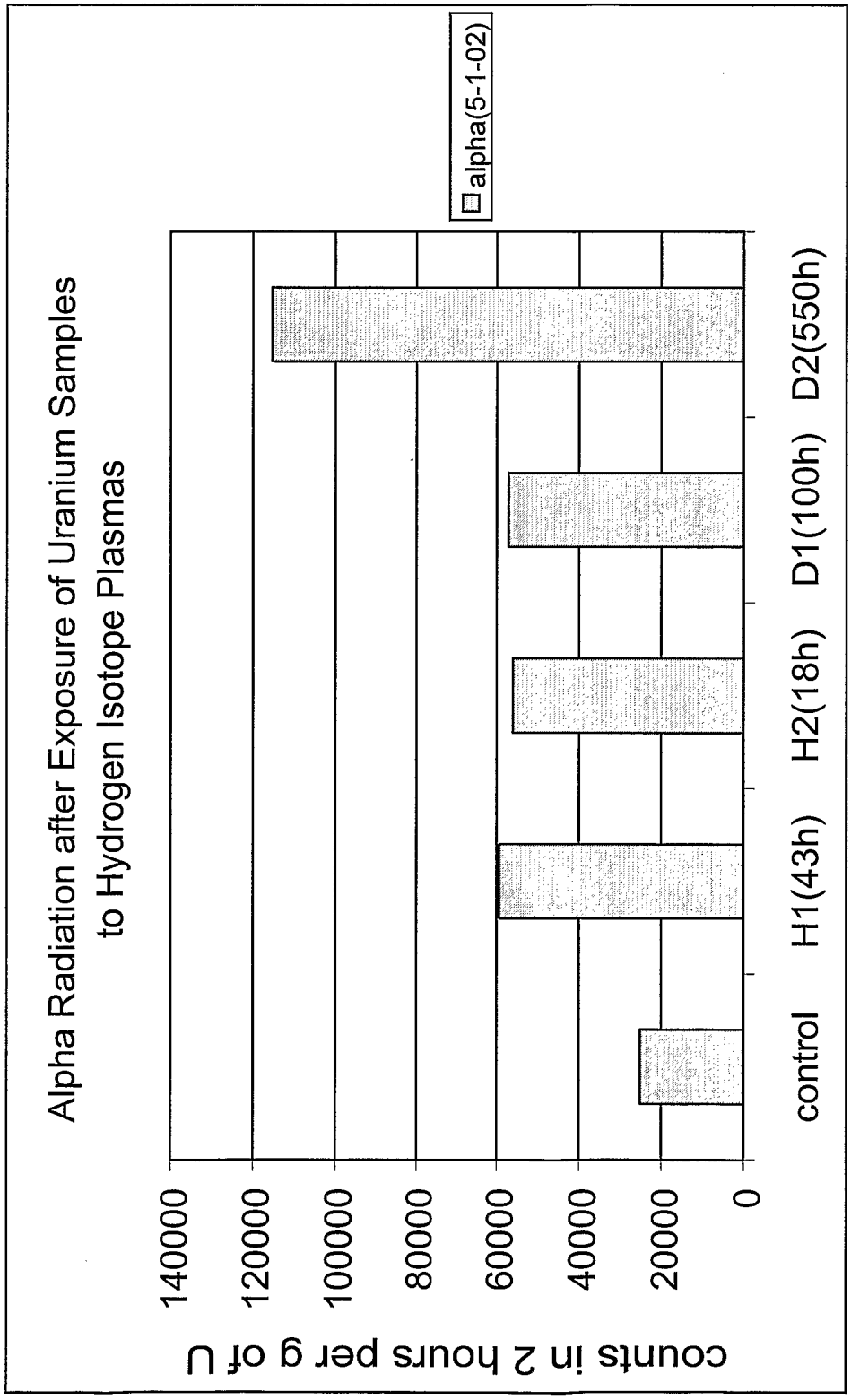

Alpha Radiation after Exposure of Uranium Samples to Hydrogen Isotope Plasmas
alpha(5-1-02)
counts in 2 hours per g of U
0
20000
40000
60000
80000
100000
120000
140000
control
H1(43h)
H2(18h)
D1(100h)
D2(550h)

FIG. 4

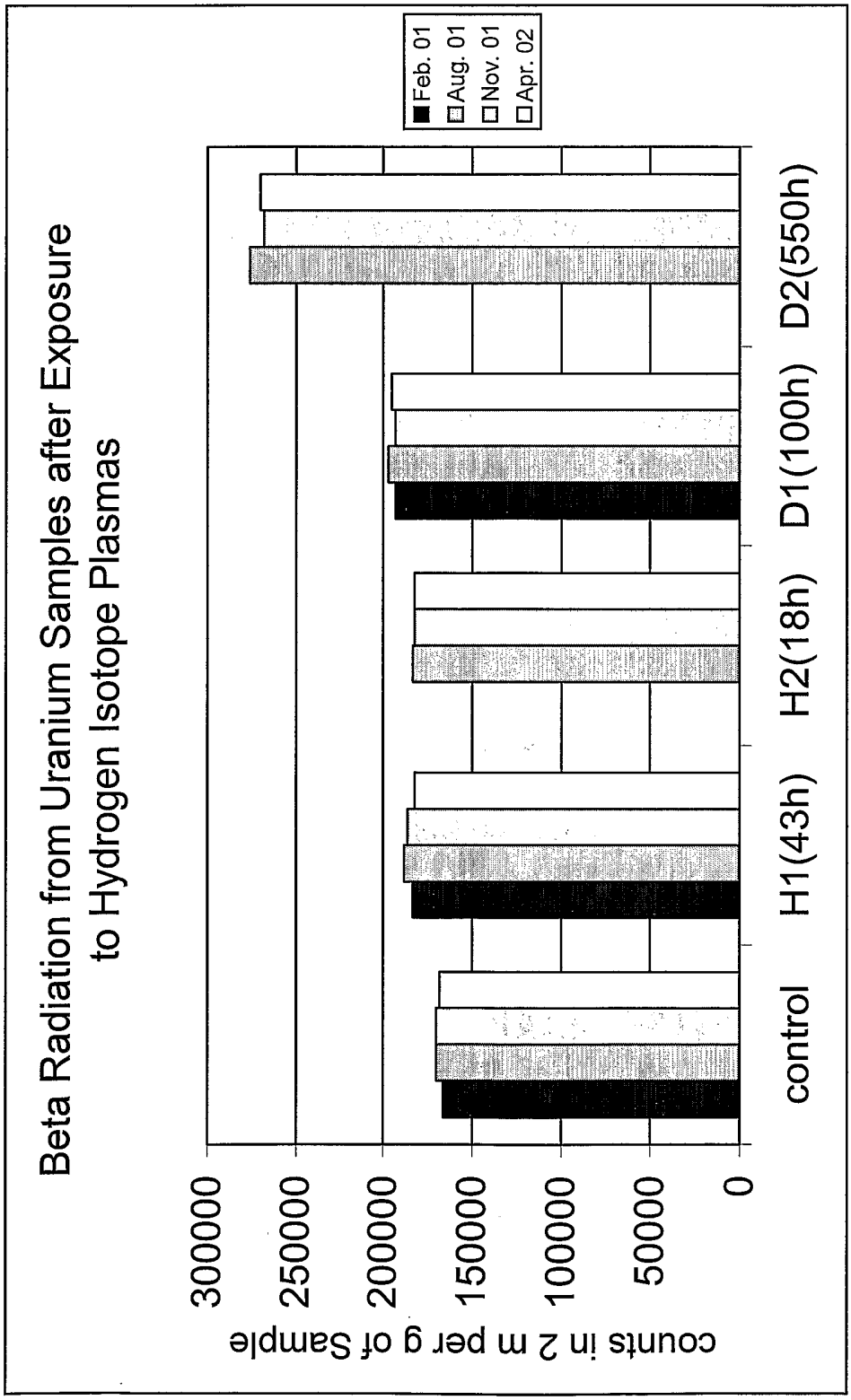

Beta Radiation from Uranium Samples after Exposure to Hydrogen Isotope Plasmas
Feb. 01
Aug. 01
Nov. 01
Apr. 02
counts in 2 m per g of Sample
300000
250000
200000
150000
100000
50000
0
control
H1(43h)
H2(18h)
D1(100h)
D2(550h)

FIG. 5

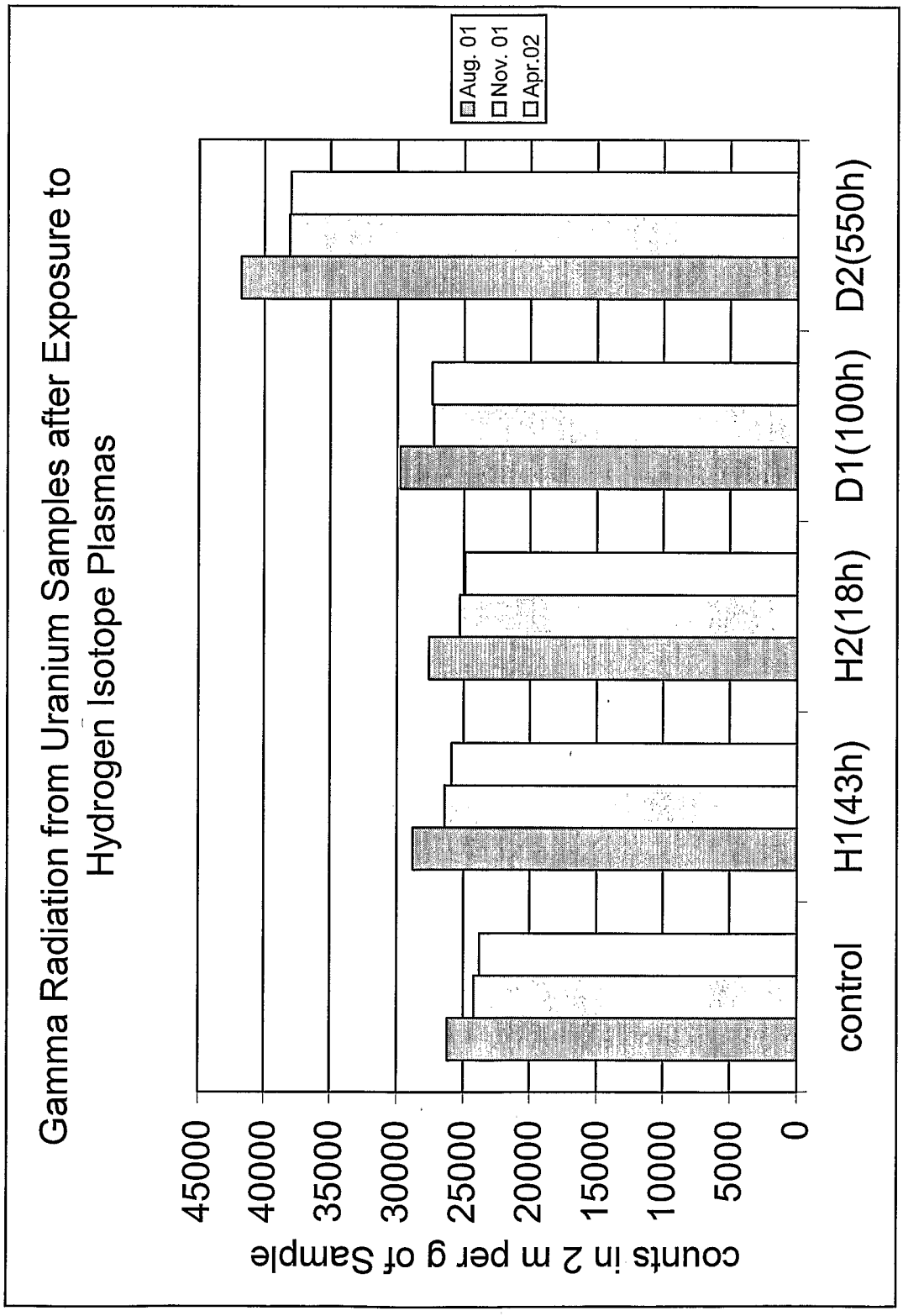

Gamma Radiation from Uranium Samples after Exposure to Hydrogen Isotope Plasmas
Aug. 01
Nov. 01
Apr.02
counts in 2 m per g of Sample
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
control
H1(43h)
H2(18h)
D1(100h)
D2(550h)

FIG. 6

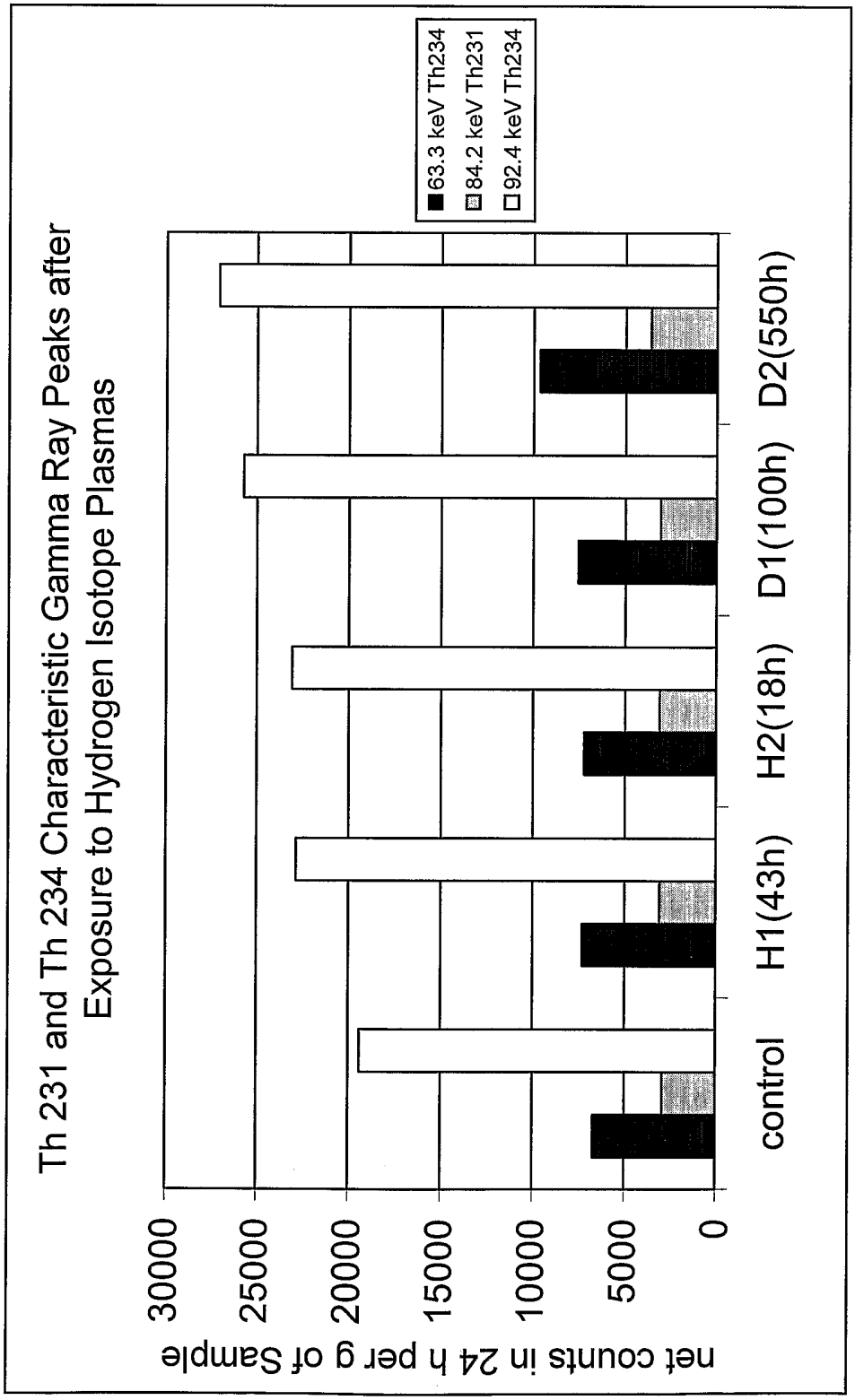

Th 231 and Th 234 Characteristic Gamma Ray Peaks after Exposure to Hydrogen Isotope Plasmas
63.3 keV Th234
84.2 keV Th231
92.4 keV Th234
net counts in 24 h per g of Sample
30000
25000
20000
15000
10000
5000
0
control
H1(43h)
H2(18h)
D1(100h)
D2(550h)

FIG. 7

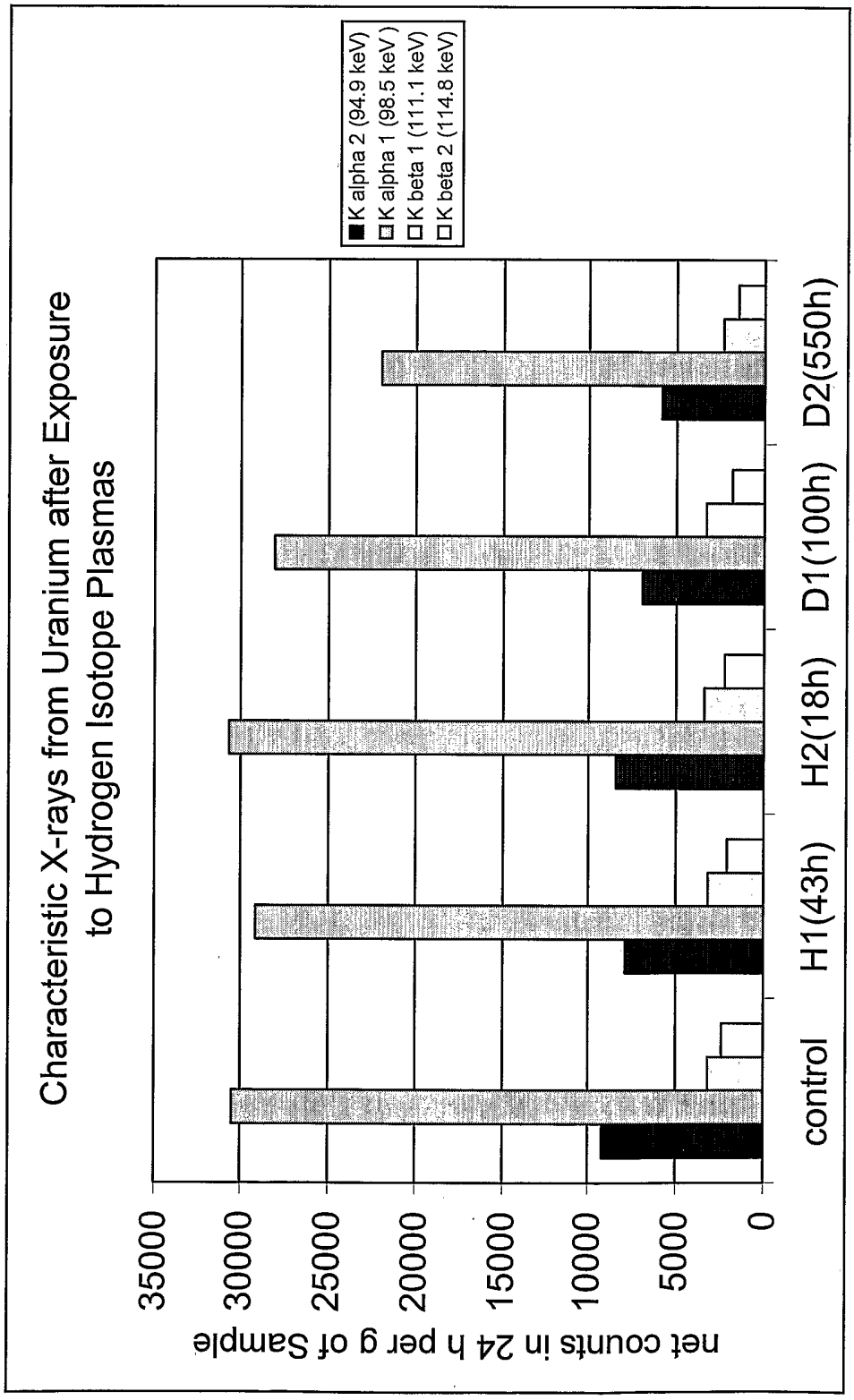

Characteristic X-rays from Uranium after Exposure to Hydrogen Isotope Plasmas
K alpha 2 (94.9 keV)
K alpha 1 (98.5 keV)
K beta 1 (111.1 keV)
K beta 2 (114.8 keV)
net counts in 24 h per g of Sample
0
5000
10000
15000
20000
25000
30000
35000
control
H1(43h)
H2(18h)
D1(100h)
D2(550h)

FIG. 8

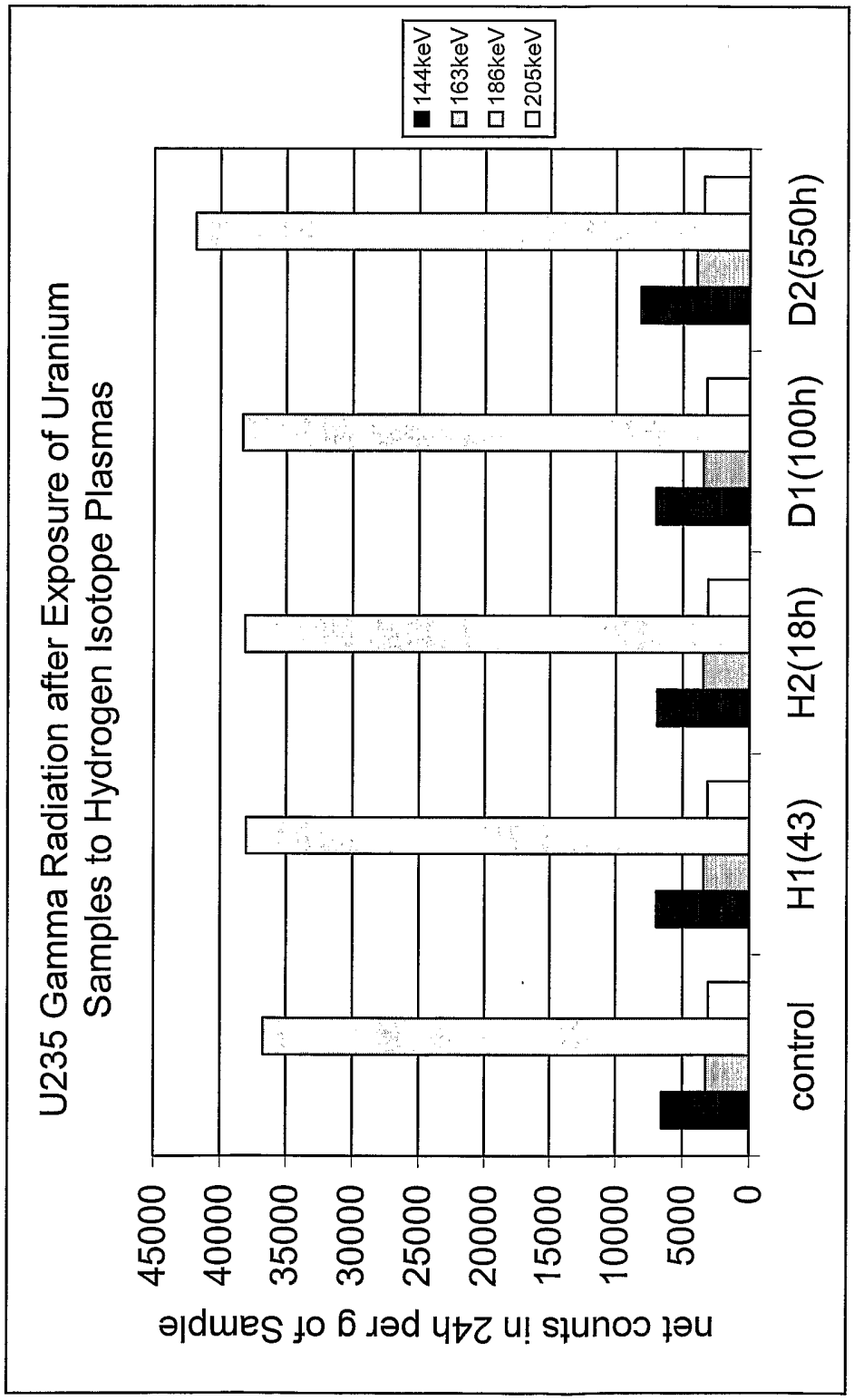

U235 Gamma Radiation after Exposure of Uranium Samples to Hydrogen Isotope Plasmas
net counts in 24h per g of Sample
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
144keV
163keV
186keV
205keV
control
H1(43)
H2(18h)
D1(100h)
D2(550h)

FIG. 9