EFFECTS OF GLOW DISCHARGE WITH HYDROGEN ISOTOPE PLASMAS ON RADIOACTIVITY OF URANIUM

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ABSTRACT

Uranium foils were attached to the cathode of a glow discharge apparatus. A plasma of either hydrogen or deuterium ions was used to bombard the uranium. The rates of alpha, beta, and gamma radiation emissions were significantly greater for the bombarded uranium than for the original material.

1. INTRODUCTION

Our previous research compared the radiation output of uranium codeposited with hydrogen (by aqueous electrolysis with nickel cathodes) to the output of the starting material, U_3O_8 [1, 2]. Radiation output was greater for the electroplated uranium. For example, the rate of alpha radiation emitted by the electroplated uranium was twice as great as the rate of alpha emission from the starting material, U_3O_8. Subsequent to our reports[1,2], we determined that the concentration of uranium in the electroplated material was 30% higher than in the starting material. Therefore, the normalized rate of alpha emission from the electroplated uranium was about 30% greater than that from the starting material. Similar results were also obtained for beta and gamma emissions.

The aim of our current research was to determine if similar effects occur in glow discharge experiments.

2. EXPERIMENTAL METHODS

A schematic diagram of the glow discharge apparatus is shown in Fig. 1. Two quartz tubes concentric with the stainless steel electrodes were sealed with gaskets against stainless steel end plates. The space between the concentric quartz tubes allows for water cooling, and there is also provision for water cooling the electrodes. Water cooling was not necessary in our experiments because the power input was very low. A molybdenum foil was held against the face of the anode with a molybdenum nut. A 19 mm diameter disc made of natural uranium (99.98% U, ~0.2 mm thick) was held against the face of the cathode with a molybdenum nut. The part of the uranium disc which was exposed to the hydrogen isotope plasma had a diameter of 13 mm. Therefore, about 47% of the area was exposed to the plasma.

Two uranium discs were exposed to hydrogen plasma, and two others were exposed to deuterium plasma. The mass of each disc was determined before and after these exposures. Before exposure the mass of each disc was about 1 g. Erosion which occurred during glow discharge reduced the mass of each disc. Glow discharge was performed with flowing gas at about 5 torr. The current during glow discharge was about 5 mA and the voltage was about 500 volts. Alpha, beta, and gamma measurements were made on each of these samples after the exposures, along with measurements on a control, which was a uranium disc from the same foil and of the same size, but which had not been exposed to plasma in the glow discharge apparatus. The alpha measurements were made with a Ludlum 43-5 alpha probe. The beta measurements were made with a Ludlum model 2000 GM counter. By placing a one mm thick aluminum plate between the detector and the sample, it was possible to detect the continuum of high energy gamma and x-rays. Gamma and x-ray spectra were obtained with an EG&G ORTEC 92X gamma ray spectrometer. Radiation measurements were made periodically over a time span of about one year. The uranium specimen with ~ 550 hour exposure to deuterium plasma and the control specimen

were examined with a scanning electron microscope (SEM) and analyzed with an energy dispersive spectrometer (EDS).

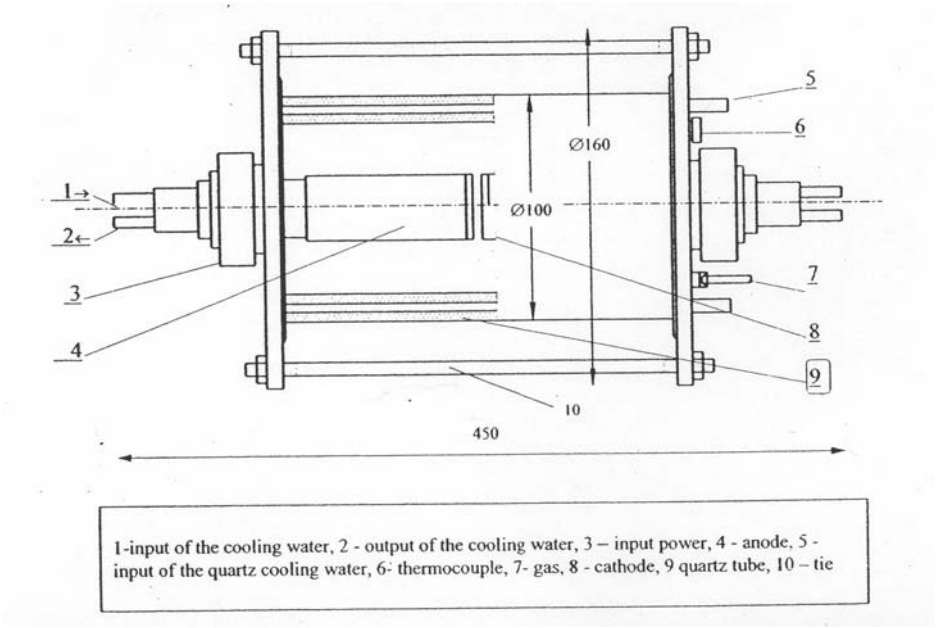


Fig. 1. Glow discharge apparatus. The dimensions are in millimeters.

3. RESULTS

Alpha radiation emitted by the control, by two samples exposed to hydrogen plasmas, and by two samples exposed to deuterium plasmas was measured on 5-2-02. The results are shown in Fig. 2, where H denotes samples exposed to hydrogen plasma and D denotes samples exposed to deuterium plasmas. The results are normalized to give counts per gram of uranium in order to take into account the differences in mass of the samples. The times of exposure to glow discharge were 43 hours in hydrogen plasma for sample H1, 18 hours in hydrogen plasma for sample H2, 100 hours in deuterium plasma for sample D1, and 550 hours in deuterium plasma for sample D2.

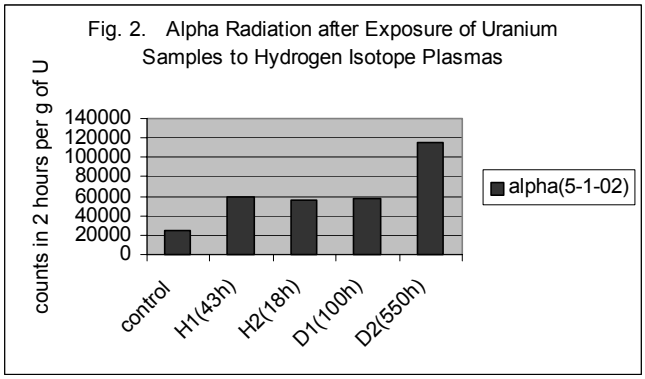
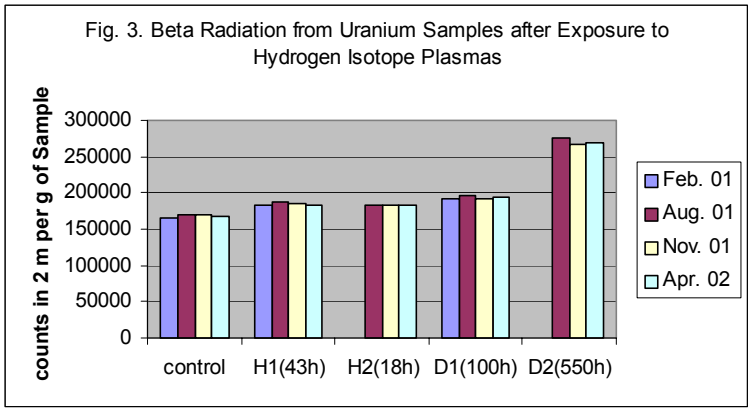


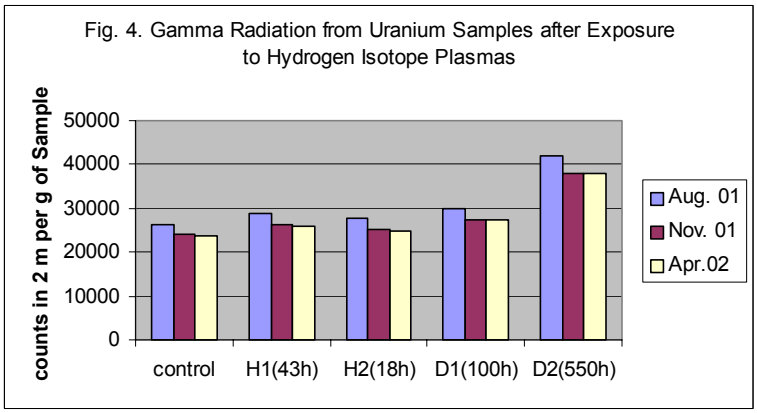
Fig. 2 clearly shows that exposure of uranium to hydrogen isotope plasmas greatly increases the flux of alpha radiation. The flux from three of the samples is almost the same and is more than twice the flux from the control. Two of these samples were exposed to hydrogen plasma and one was exposed deuterium plasma. The flux from

the fourth sample, which was exposed to deuterium plasma for ~ 550 hours, is more than four times as great as the alpha flux from the control..

Fig. 3 shows the results of beta radiation measurements taken 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 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.



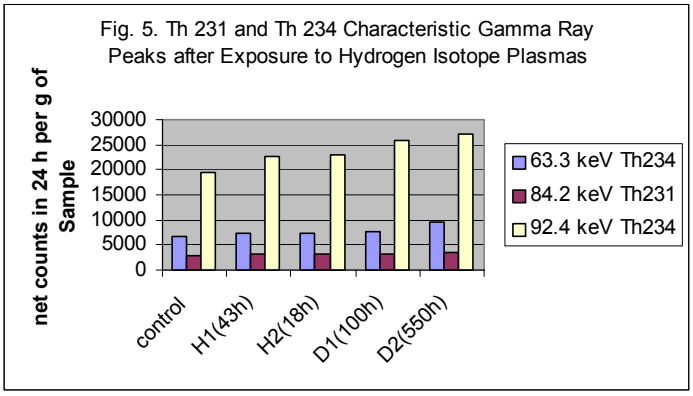
The next data set was obtained by placing a one mm thick aluminum plate between the Geiger counter and the specimen in order to block beta radiation. The results are given in Fig. 4.



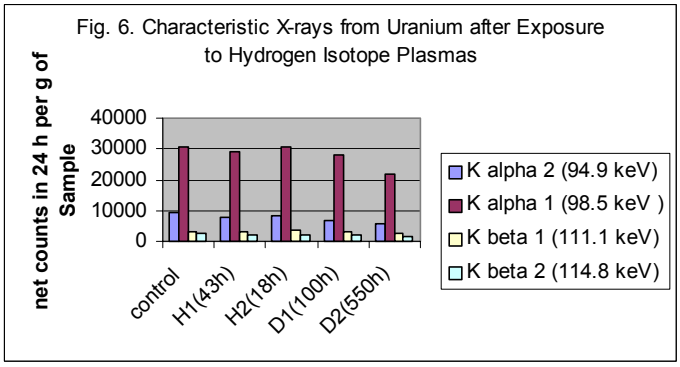
It is thought that the Fig. 4 differences in radiation obtained for each sample over the time span from August 01 to April 02 was caused by slight differences in geometry between the samples, the aluminum plate, and the detector. Here again, all of the samples exposed to hydrogen isotope plasmas have greater fluxes of radiation (high energy gamma and x-rays) than the control. The amounts of the increases are about 10% for H1, 5% for H2, 14% for D1, and 59% for D2.

Eleven of the most prominent characteristic peaks in spectra obtained with the gamma ray spectrometer were analyzed to determine possible correlations with the other data. The first peak in the gamma ray spectra, 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 intensities of these peaks obtained from the control and from the four reacted samples

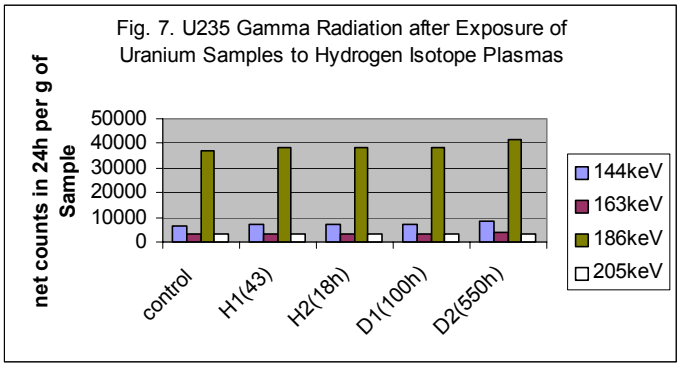
are shown in Fig. 5. The net count for every peak from the reacted samples is greater than for the corresponding peak from the control. These differences are most apparent for the 92.4 keV peaks. The intensity of this peak for H1 was 18% greater than the control, for H2 it was 19% greater, for D1 it was 33% greater, and for D2 it was 39% greater.



The next four most intense peaks 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 Kβ4 at 114.8 keV. The intensities of these peaks for the control and for the four reacted samples are shown in Fig. 6. Here it is seen that all of the Kα2 and Kβ4 peaks for the reacted samples are **less** intense than the corresponding peaks for 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. It should be recalled that 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 varied results for Kβ1 may be caused by overlap with some as yet unidentified peak. The uniformly large decreases in these characteristic x-ray peaks of uranium for sample D2 suggests that this sample has less uranium per unit mass than the control. The same conclusion may also apply to the other three reacted samples.



At higher energy in the gamma ray spectra, the next four prominent peaks are gamma rays emitted by U235. Fig. 7 shows the intensities of these peaks for the four reacted samples and for the control. Although the differences are not as great as those in Fig. 5, here again the gamma ray emissions are greater in every case for the reacted samples H1, H2, D1, and D2 than for the control. Again, 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.



Specimen D2 was characterized by SEM and EDS after cathodic bombardment with deuterium ions in the glow discharge plasma. Fig. 8 is a low magnification photograph of a portion of this sample. On the left side, the relatively smooth arc represents the area of the sample which was shielded from the plasma by the molybdenum nut which held the uranium sample in contact with the stainless steel cathode. To the right of the protected area, the topography is very rough and varied in contrast. A great deal of erosion occurred in the exposed area, resulting in loss of nearly half the mass of the sample. The control sample was also examined in the same way. The EDS spectra from the control sample did not contain evidence of thorium, but thorium was detected in many spectra from D2. A typical analysis gave 1% thorium with a standard deviation of 0.3%.

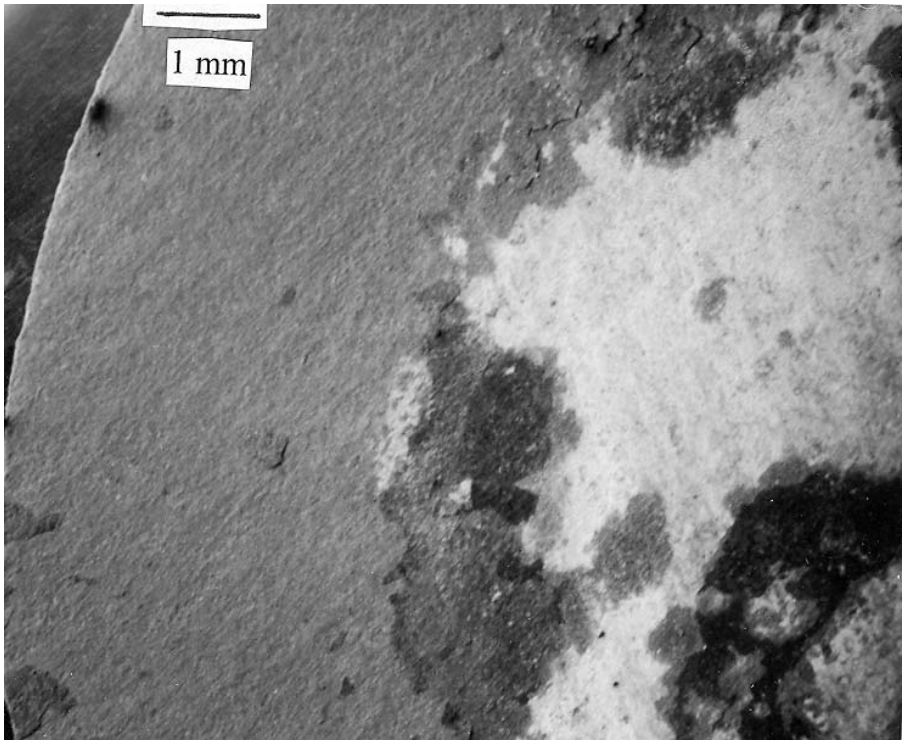


Fig. 8. SEM photograph of uranium sample D2 after exposure to deuterium plasma for about 550 hours. The relatively smooth rim on the left was protected from exposure to the plasma by a molybdenum nut which fastened the sample against the cathode.

4. DISCUSSION

The alpha, beta, and gamma data presented above clearly indicate that the interaction of uranium with hydrogen isotopes during glow discharge results in shortening of the decay time of uranium. The data in Fig. 2 show that glow discharge for 18 hours with hydrogen plasma reduces decay time by more than a factor of two. Decay time was reduced by more than a factor of four when uranium was exposed to deuterium plasma for 550 hours. It should be emphasized that only about one-half of each sample was exposed to plasma and only on one side. It should be expected that larger effects would have occurred if the entire surface of each sample had been exposed to plasma on both sides. Similar results were obtained in our previous research on codeposition of hydrogen and uranium from a light water electrolyte [1, 2].

Bush [3] found evidence for beta decay of ^{40}K to ^{40}Ca. His results suggest that the half life of ^{40}K was greatly reduced from the normal 10^9 years. Change over a short time span in the quantity of ^{107}Pd, which has a half life of about 7×10^6 years, has also been reported [4]. This also suggests a great reduction in half life.

5. REFERENCES

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6. ACKNOWLEDGMENT

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