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Research Article

Neutron Emission from Cryogenically Cooled Metals Under Thermal Shock

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Abstract

During the summer of 1991, intense neutron bursts were observed after temperature shocking titanium chips which had been saturated with deuterium gas. The titanium chips were cooled and loaded with deuterium at 77 K and then rapidly heated to 323 K. The rapid heating produces a large pressure increase inside the crystalline lattice of the host metal. An Event Timer/Counter (ETC) card was designed and developed which counted and kept a time distribution of the neutron pulses as they occurred from a helium-3 neutron counter embedded in a paraffin moderator [1]. The experiment produced copious neutron counts. During one cooling and heating cycle, over 2 million neutrons were counted over a 5 min time period. In subsequent cooling and heating cycles using the same titanium chips, significant neutron bursts were observed with diminishing counts after each subsequent cycle. This paper will discuss the 1991 experiments and the status of ongoing experiments. .

Keywords: Deuterium, Neutrons, Phase Change, Surface preparation, Thermal shock, Titanium

1. Introduction

Reports indicated that low level neutron bursts occurred in titanium [2] and palladium [3]. Both experiments, designed to look at neutron production from deuterium fusion, created a highly stressed lattice prior to the observed neutron bursts. Experiments were designed by our group to take advantage of a potentially large pressure build-up in the crystalline lattice of metals created by phase changes during a thermal shock from liquid nitrogen temperature (–190^{o}C) to 50^{o}C. The palladium hydride phase diagram is distinctly different from that of titanium hydride. Palladium hydride retains its face centered structure throughout the few phase changes it experiences. The changes of phase are simply a distortion of the crystalline lattice from face centered cubic to face centered tetragonal. Information on titanium is not as plentiful as with palladium but its hydride phase diagram is much more complex, with phases going from hexagonal close pack, to face centered cubic, to body centered cubic, with intermediate phases being combinations of these. Titanium, containing a high level of hydrogen, will have yet another phase transition between 77 and 300 K which can achieve greater than 60% atomic hydrogen loading at cryogenic temperatures [4]. The experiments were based on an interesting thermal

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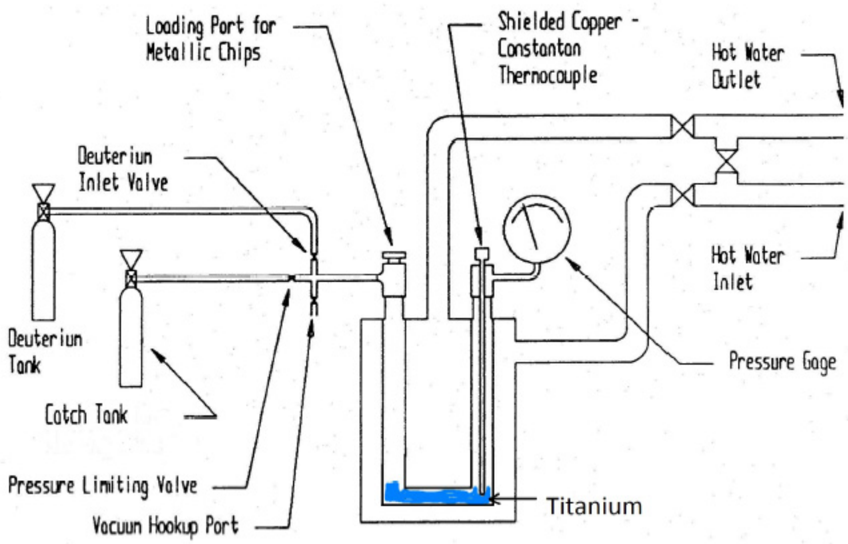

Figure 1. Sketch of the testing chamber used for the pressure shock experiment. The pressure gauge was a bourbon tube with a 1000 psi maximum and an accuracy of 3%. The catch tank was evacuated and used to capture the D_2 gas from the test chamber for analysis. The hot water inlet and outlet were used to thermally shock the samples.

shock scenario. For example, if palladium were saturated with hydrogen at liquid nitrogen temperature (–199^{o}C), the atom ratio of H/Pd would approach 1. If the temperature were quickly raised above 25^{o}C, the internal pressure would exceed 14 kbar. Titanium hydrides have not been as extensively studied as palladium hydrides, and low temperature charts are not available, but given the complex phase structure of titanium, the generation of high pressures under thermal shock was thought to be feasible.

2. Experiment

The initial cryogenic thermal shocking experiments used 42 g of 99.5% pure titanium sponge (from Alfa Aesar) which was broken into 1/8 inch or smaller chips by hand in a glove box under argon overpressure and then placed in a glass jar. The jar was sealed and agitated using a paint shaker for several minutes and the pieces further fragmented. A double-walled chamber was built with the inner container made of high-pressure copper tubing bent into a U-shape and an outer chamber made of 304 stainless steel. A T-joint was placed on the ends of the U-shaped copper tubing. On one side there was a loading port at the top of the T (where the titanium chips were loaded) and a gas manifold on the side. On the other part of the U a thermocouple was inserted and a pressure gauge (Fig. 1).

The test chamber was designed to be moved from a liquid nitrogen bath and placed in the center of a neutron

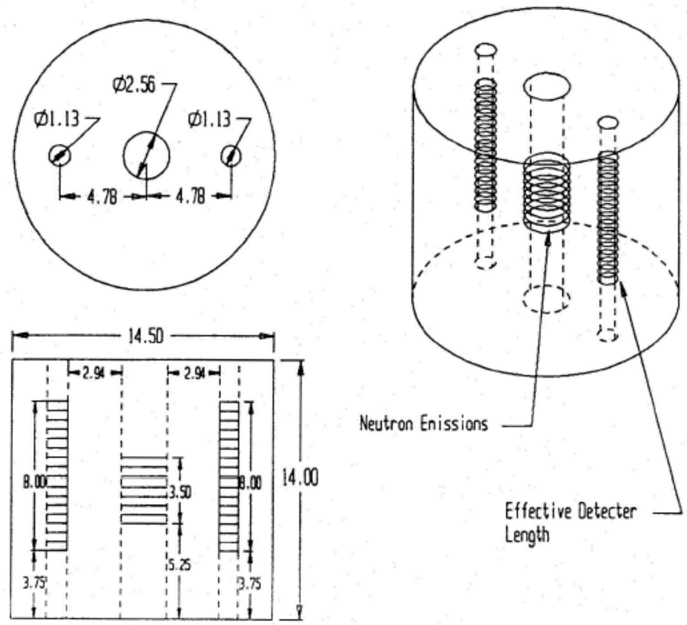

Figure 2. A solid paraffin block was fitted with two helium-3 detectors in the position labelled as "effective detector length." The dimensions are in inches.

counting chamber, using two helium-3 detectors, built to detect low-level neutron bursts (Fig. 2).

The helium-3 detector signals were amplified, shaped and then sent to the ETC card in a Macintosh II computer and to a decade counter. The ETC was designed to achieve one microsecond resolution and a 32,000 count storage capacity [1]. It was designed to run unattended until its storage capacity was filled and then shut down. Data was then transferred to a storage file on the hard drive. Afterwards the card could be cleared and restarted. The decade counter simply accumulated counts over a set period of time. Once it reached its capacity of 1 million counts, it reinitialized and started the count cycle over. Given that all experiments had shown low level bursts, it was believed that the 32,000 count capacity of the ETC and the 1 million count capacity of the decade counter were sufficient.

3. Results

Timing of the various measurements was complicated. The computer clock was started by software when the test chamber was placed in the neutron counter. The temperature and pressure were timed and observations recorded by a student with a chronograph watch. The decade counter and ETC were located in a Faraday cage and the signals were brought into the room with BNC connectors. The student timed and recorded the decade counter data with a chronograph watch. All times were synchronized to a time zero corresponding the ETC start time.

A pressure loading with argon was run as a control where the test chamber was pressurized to 200 psi argon and

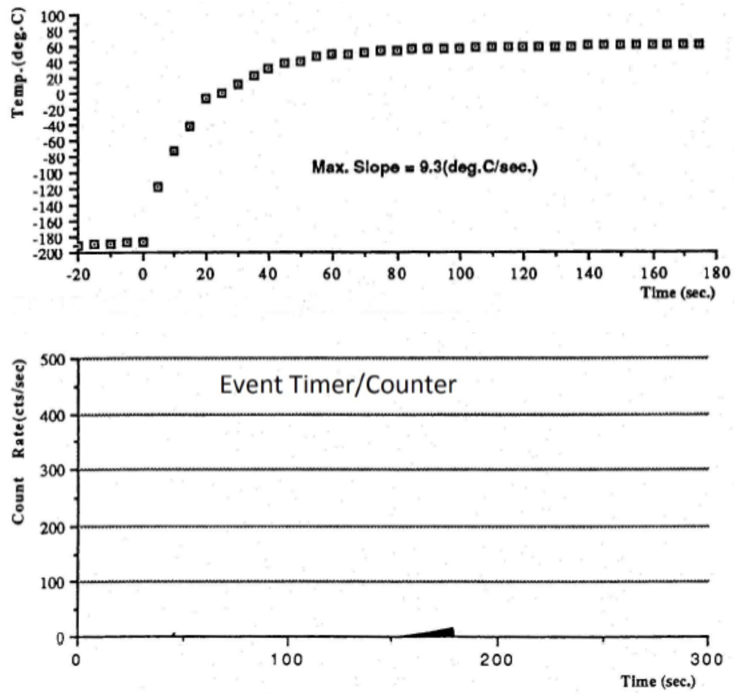

Figure 3. Thermal shocking using argon. ETC and decade counter show 45 counts in 240 s.

then cooled in the LN2 bath until pressure and temperature reached equilibrium. Then the test chamber was loaded into the neutron counting unit and hot water injected into the test chamber. The results from this test are shown in Fig. 3. The ETC data is plotted below the temperature data. The decade counter showed count rates consistent with the ETC and matched the results from prior extensive background counting runs (where the background was on the order of 0.15 counts per second).

As shown in Table 1, a procedure for loading the titanium with D_2 was developed to limit exothermic heat from the creation of titanium deuteride. The D_2 pressure was slowly increased to 200 psi (the valve was closed at 200 psi) while simultaneously running hot water (at 45^{o}C). When the temperature stabilized, the water was drained and the test chamber was slowly lowered into an Liquid Nitrogen (LN2) bath on July 20, 1991 at 01:54. As temperature dropped, the D_2 pressure fell below 50 psi and at this point the D_2 pressure was increased to 100 psi and the valve closed. The D_2 pressure in the test chamber dropped and the temperature increased. At 310 min the decision was made to take the test chamber from the LN2 bath and place it in the neutron counter (even though pressure was still dropping and temperature increasing slowly). At 313 min, hot water was injected into the chamber causing the temperature to rise (Fig. 4). The hot water circulated for 15 min and then was shut off. The test chamber stayed in the neutron counting chamber for three days with the ETC and decade counter running. In the first cycle the neutron counts were at background level.

Since the test chamber was still showing changes in pressure and temperature when the test chamber was removed from LN2 during the first cycle, we thought that the chips had not been fully loaded. On July 23, 1991 at 18:42, the test chamber was then cooled to –186^{o}C and the D_2 pressure was held at 50 psi for 160 min. The ETC was started and at 163 min, the test chamber was placed in the neutron detection chamber and the hot water flow started. Due to a freezing problem, the water flow was restricted and the rate of temperature change was reduced. As soon as the test chamber

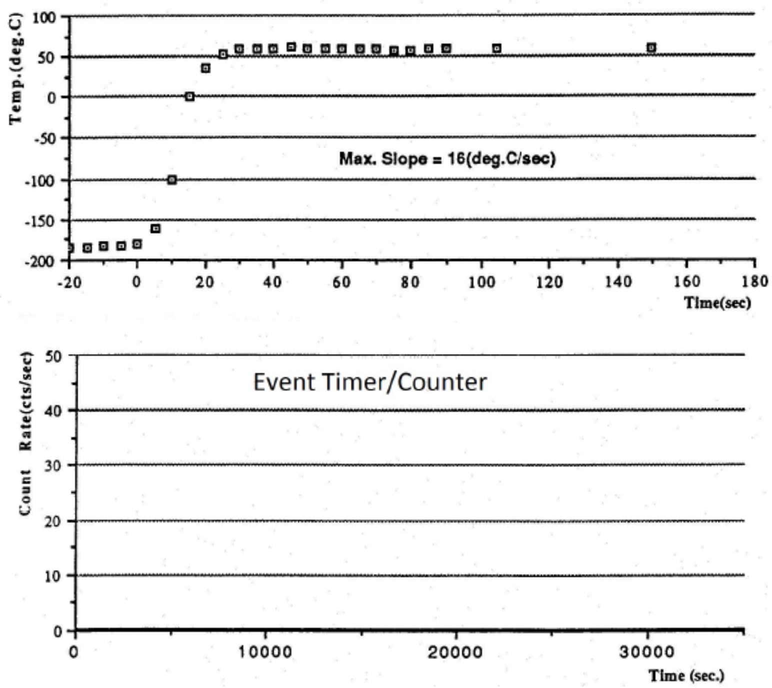

Figure 4. In the first thermal shock cycle, the ETC and decade counter were consistent showing no neutrons counts above background.

was in the neutron counting chamber, neutron counts were rapidly building up on both the ETC and the decade counter. The ETC's storage capacity was quickly saturated. The ETC had to be manually restarted about 42.8 s after saturation.

It again saturated in about 0.11 s. A computer glitch did not allow a restart after the second saturation. In the meantime the decade counter had exceeded its one million count capacity at least twice (the student recording decade counter data by hand was also the student who restarted the ETC three times likely missing other decade counter saturation events- the counts that were observed were at a consistently high count rates). After 5 min, it was decided to put the device back into the LN2 bath.

The results from the second temperature cycle had a minimum 2,486,500 neutron counts during the five-minute period. The detector efficiency was ~4% so this would correspond to a minimum of 62,163,000 neutrons produced. A more accurate indication of neutron counts was only possible during the two brief periods when the ETC was running and saturated. Figure 5 shows the two points in time where the ETC was saturated in about ~0.11 s. This data indicates 290,000 counts per second.

A third cycle was started by putting the test chamber in a LN2 bath for 35 min (cooled to –190^{o}C). On July 23, 1991 at 19:25 the chamber was allowed to warm by natural convection. Everyone left for the night (due to fatigue) and returned 17 h later. The decade counter showed 50,883 counts and the ETC had saturated. The decade counter could have recycled itself multiple times during the event shown in Fig. 6 so the total count is at minimum 50,833, but could have orders of magnitude more. One small neutron burst occurred within 10 s of the convective warming process and a much larger burst which saturated the ETC occurred at 47,715 s into the cycle.

A fourth cycle was started on Wednesday July 24, 1991 at 17:20. The test chamber was cooled to –190^{o}C for 190 min and the D_2 pressure held at 50 psi. The ETC was started and the test chamber put into the neutron counting

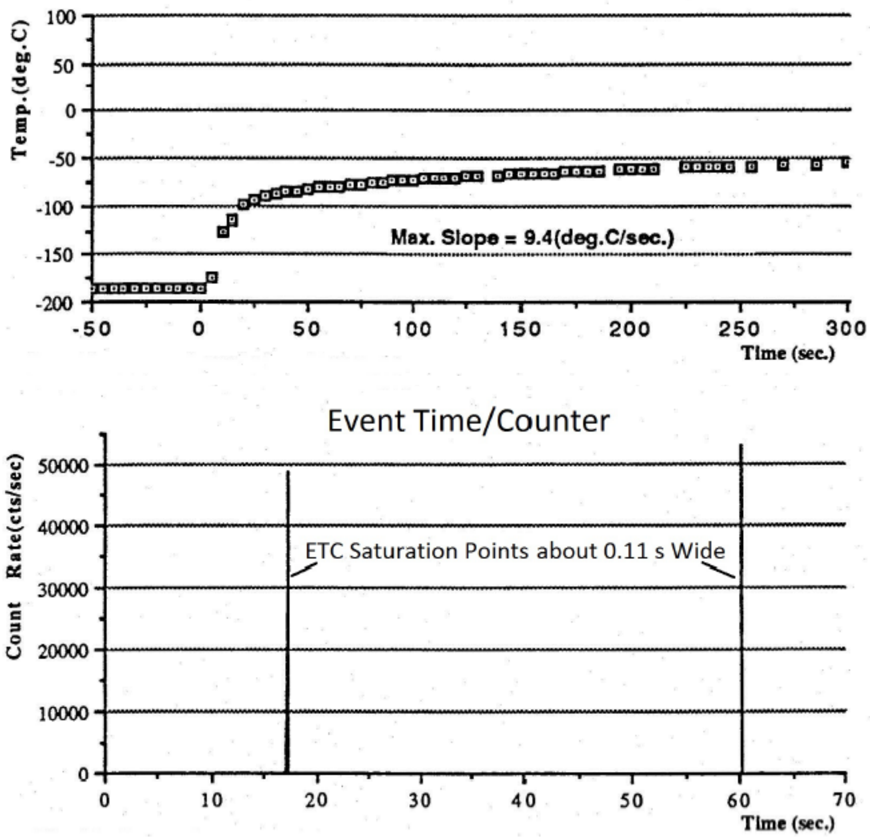

Figure 5. Second cycle neutron count rate.

chamber and at 20:25 hot water was injected. A neutron burst was seen about 10 s after the start of the heating cycle (Fig. 7).

At this point we collected the D_2 gas and titanium samples to check for tritium content. One hour and 26 min after removing the titanium, 9.6 g of the sample was placed in a Tri Carb 1600 TR Liquid Scintillation Analyzer. The minimum detectable level of tritium for this unit was 2.9×10^{-6} μCurie. The test sample yielded a reading of 12.8×10^{-6} μCuri. A control sample of the titanium yielded a reading of 11.9×10^{-6} μCuri. There was a difference of 0.9×10^{-6} μCuri.

4. Discussion and Conclusions

Cryogenically cooled metals under thermal shock yielded promising results. Three cooling and heating cycles using titanium yielded neutron counts significantly above background. The ETC was able to pinpoint when the bursts occurred and the count rate at the time of the burst. In the second cycle, a sustained burst of neutrons were observed that twice saturated the 32K storage limit of the ETC before a software failure and a decade counter at least twice exceeded it's one million count capacity during the five minute period. The neutron production might have been sustained longer had we not put the test chamber back into the LN2 bath in order to try to stop the reaction. At minimum, this event

Table 1. Data showing the procedure used to saturate the titanium with deuterium. D_2 pressure was slowly increased to 200 psi at which point the valve was closed. When D_2 pressure dropped below 50 psi, the valve was opened to increase it to 100 psi and was closed.

Time (min)	Pressure (psi)	Temperature (°C)
0	40	28.9
20	40	50.6
45	60	45.0
65	80	45.0
80	100	45.0
85	120	45.0
90	140	45.0
95	160	45.0
100	200	45.0
105	230	98.3
115	225	91.6
125	215	45
135	Drain water and slowly lower in LN2 bath	
140	200	–5.0
150	180	–72.0
155	140	–117.0
160	115	–136.0
180	45	–182.0
190	100 (add D_2)	–190.0
280	100	–188.0
300	60	–186.0
310	55	–184.0

yielded >2,486,500 neutron counts (the student who was observing the decade counter was busy trying to restart the ETC and missed additional cycling of the decade counter). In addition the ETC was saturated twice. The saturation

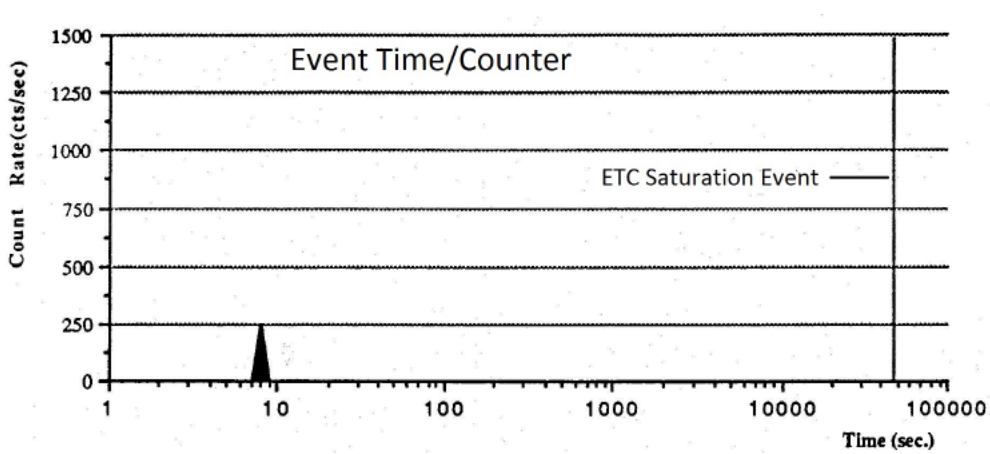

Figure 6. Event counter results from the third cycle.

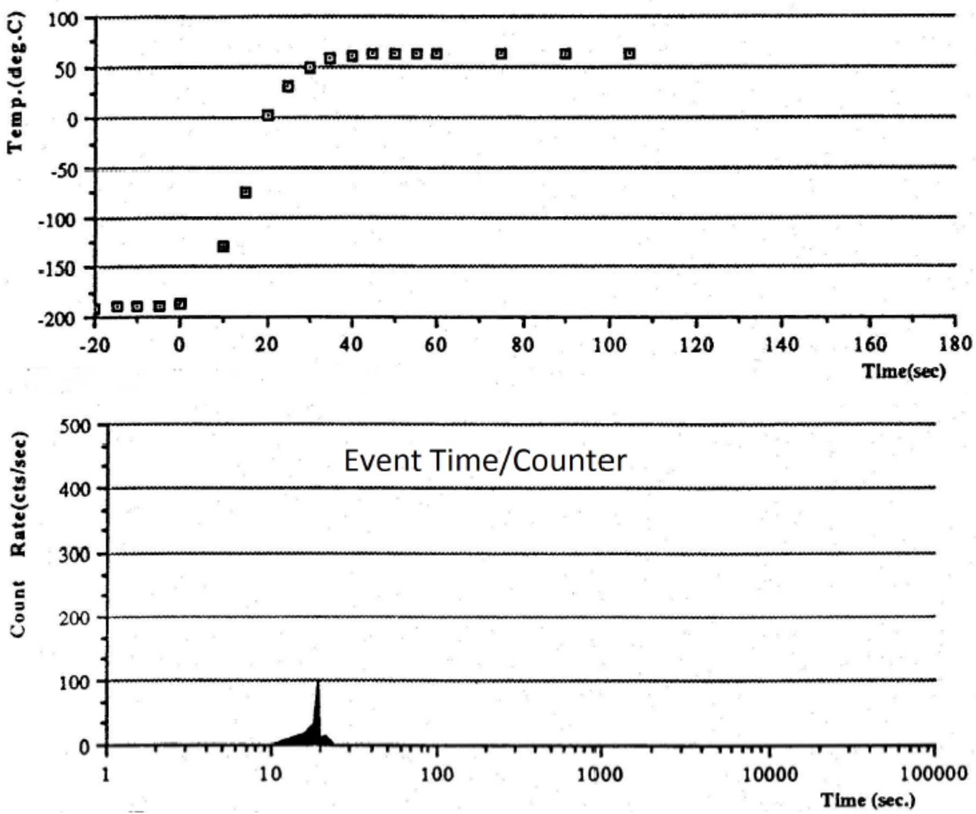

Figure 7. Fourth heating cycle.

time was 0.11 s which indicated a count rate of 290,000 counts per second. Our goal was to test other metals capable of forming hydrides (e.g., palladium). However, the experiments were discontinued due to circumstances beyond our control. In the 22 years since this work, others groups have seen neutron bursts in various metal hydrides. This particular experiment is unique in both magnitude and duration of neutron production.

As part of the Sydney Kimel Institute for the Nuclear Renaissance, a continuation of the cryogenically cooled metals under thermal shock experiments are underway. A team is developing a next generation testing system—some of which will be reported at future meetings and in future publications.

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