

SEARCH FOR CHARGED - PARTICLE d - d FUSION PRODUCTS IN AN ENCAPSULATED Pd THIN FILM

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ABSTRACT

Motivated by reports by Fleishmann and Pons and also Jones *et al.* of nuclear fusion occurring at room temperature, we attempted to look for charged particle reaction products from d-d fusion in a deuterated palladium thin film. A silicon nitride encapsulated palladium thin film (340 nanometers thick and one square centimeter in area) was fabricated on top of a semiconductor particle detector and implanted with an 80 keV D_2^+ beam. The purpose of the nitride cap was to prevent deuterium from diffusing out or from being sputtered away during implantation. The detector temperature was maintained below 200 K in order to reduce pressure on the cap. During the first run of this experiment, after the ion implanter had been turned off, apparent charged particle guises as well as bursts of activity in two nearby Geiger counters were observed with the film loaded to a nominal 150% deuterium-to-palladium ratio and a 1.3% dose of 6Li . No spectrum was obtained because of equipment malfunction. In a second run no apparent charged particle pulses were observed, but a record of the neutron flux due to induced fusion during implantation suggested that the nitride cap had failed. More experimental runs are expected in the near future.

INTRODUCTION

Reports by Fleishmann and Pons ¹ and Jones et al ² have suggested the possibility of deuteron-deuteron (d-d) fusion at room temperature within the bulk palladium electrode of an electrochemical cell having a heavy water electrolyte solution. The heat measured in such a cell by Fleishmann and Pons was greater than what could be accounted for by a chemical reaction. Jones and his co-workers measured a low neutron flux from a similar system. Both of these experiments and others ^{3,4} as well looked for heat and/or neutrons but not for the charged-particle products of the dominant d-d fusion reactions



(A third reaction, $d + d \longrightarrow {}^4\text{He} + \gamma$, is possible but only at a rate which in normal circumstances is extremely low.) It is difficult to detect these charged particles in an electrochemical cell because they interact strongly with the bath. For example, a 3 MeV proton has a range of only about 0.15 mm in water.

The purpose of our experiment was to search for charged particles produced by d-d fusion within a thin film of palladium which had been deposited on a semiconductor particle detector and implanted with deuterons to a D / Pd ratio exceeding 100 %. The palladium film was encapsulated with a thin layer of silicon nitride in order to prevent the deuterium from diffusing out or from being sputtered away during the implantation process. {Implantation through a capping layer is a standard technique used in the manufacture of integrated circuits.} The device was maintained at a temperature below 200 K in order to reduce the pressure on the cap.

DEVICE STRUCTURE AND FABRICATION ⁵

Figure 1 shows the structure of the devices used in the experiments reported in this paper.

The substrate is a one-inch diameter Quantrad 500-PN double-diffused detector made from nearly intrinsic p-type silicon with resistivity of 7,000 to 10,000 ohm-cm. It has a doped p+ back contact layer and a doped n+ entrance layer with a maximum depth of 0.3 microns. This type of detector is often called a *pin* diode.

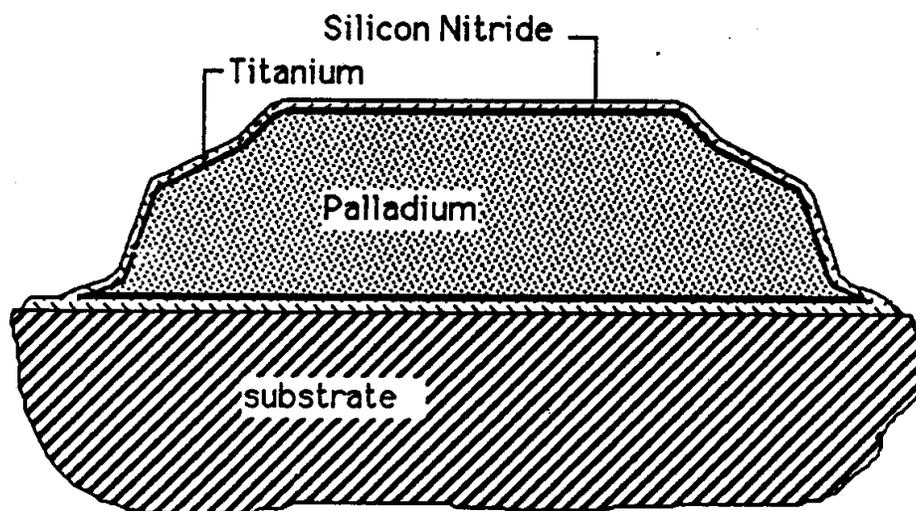


Figure 1: Cross section of thin film device.

The nitride encapsulation serves at least two purposes: it protects the deuterated palladium film from being sputtered by the deuterium ion beam during the implantation process, and it prevents the implanted deuterium from diffusing out. For sintered silicon nitride, the diffusion constant for tritium D_T is on the order of 10^{-39} cm²/sec at 300 K ⁶. The diffusion constant for deuterium D_D is $1.23D_T$. This makes the nitride virtually impermeable to deuterium.

The encapsulated palladium thin film was fabricated in five steps. Deposition of each layer was done through one of three 0.040 inch thick stainless steel shadow masks. These masks provided sloped sidewalls for easy step coverage by subsequent layers, and they eliminated the need for photolithography, which is a more complex process.

In the first step, the lower layer of silicon nitride was deposited through mask No.1 in a home-built sputtering system at the Stanford University Center for Integrated Systems. This 100 nanometer film has a diameter of 1.70 cm and an area of 2.26 cm². The reactive sputtering

process used a gas mixture of Ar:N₂ at a ratio of 1:2 and an rf power of 250 Watts. The system was pumped down to 3×10^{-6} Torr before establishing a process pressure of 10 mTorr. No temperature control was used. The nitride films have a density of 2.70 gm/cm³ with N/Si = 1.25 and an index of refraction of 1.95.

Reactive sputtering proved to be superior to chemical vapor deposition (CVD), another common method for depositing silicon nitride films. The CVD process could not be used because of the high hydrogen content in the process gases (ammonia, NH₃, and silane, SiH₄). Such a process would introduce unwanted hydrogen into the palladium film. The temperatures involved (about 800 °C) also can alter the doping profiles in the semiconductor detector substrate.

In the second step a thin layer of titanium was deposited through mask No. 2 at the San Francisco State University Thin Film Laboratory in a Varian VI-360 single-crucible 6 kW electron beam evaporator. Electron beam evaporation was used because of the high purity of the deposited films. Good step coverage was achieved with the domed, canted planetary wafer holder available in the vacuum system. The pressure during deposition was maintained below 10^{-7} Torr. The titanium "glue layer", which is a common structure in microchip fabrication, was used because palladium films do not adhere to silicon nitride. The 20 manometer titanium film has a diameter of 1.25 cm and an area of 1.23 cm².

The palladium film was deposited next in the same e-beam evaporator. Mask No. 3 defined the one cm² film. The process began when the base pressure reached 10^{-7} Torr. The palladium, which was in an alumina crucible liner, was preheated for two minutes and then deposited at a pressure below 10^{-5} Torr. The film thickness is 340 nanometers. The volume of the Film (3.40×10^{-5} cm³) contains 2.31×10^{18} palladium atoms. Thus the dose for 100% loading is 2.31×10^{18} D/cm² or 1.15×10^{18} D₂/cm².

Another titanium glue layer was deposited, again through mask No. 2, on top of the palladium film. This glue layer provided adhesion to the final nitride layer.

There was no temperature control in any of the above depositions.

With mask No. 1 in place the final nitride layer was reactively sputtered to a thickness of 100 nanometers.

CALCULATED ENERGY SPECTRUM

Fabrication of the palladium thin film directly on top of the particle detector allows very efficient collection of any particles which may be emitted from the film. However, the varying trajectories of the particles in the palladium film distort the energy spectrum by causing peaks to be smeared out. Figure 2 shows the log of the number of counts as a function of energy deposited in the semiconductor detector with the assumption that the particles are emitted isotropically throughout the palladium film.

RANGE CALCULATIONS

The implantation energy was selected to be high enough to ensure that 95% of the incident ions stop in the palladium rather than in the encapsulation layer and low enough to ensure that one third of the film suffers no implant damage. Lattice damage could in principle interfere with any possible cold fusion process by distorting the palladium crystal structure.

Range calculations were done using TRIM-90⁷. The optimum implant energy was found to be 40 keV per deuteron. As shown in Figure 3, the total range in the thin film device is 244 nm. This corresponds to a range of $R_p = 124$ nm into the palladium film with a standard deviation of $\Delta R_p = 52.5$ nm.

An 80 keV D₂⁺ ion beam was used rather than a 40 keV D⁺ beam, because the heavy water source produced higher D₂⁺ current than D⁺ current, and the D₂⁺ beam could be focused more easily. Note also that the D₂⁺ beam provides twice as many deuterons per microAmp as the D⁺

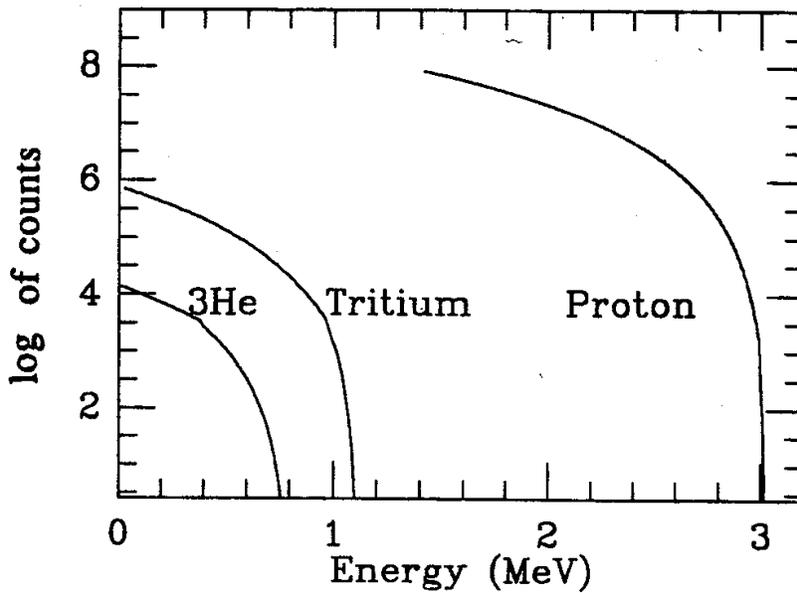


Figure 2: Calculated energy spectrum. RANGE

beam. Each incident 80 keV D_2^+ breaks into two deuterons of equal energy (40 keV) as it penetrates the device.

INDUCED FUSION

An deuterium beam incident on a deuterated target will induce fusion. A one milliAmp 40 keV D^+ beam incident on a target with 10^{22} D/cm³ generates a neutron dose of 8 mrem/hr at one meter⁸. This calculation was done by considering a uniform distribution of target deuterons throughout the film. Results of the calculation are within a factor of two or three of experimental results from Los Alamos National Laboratory. In this experiment the neutron dose should be

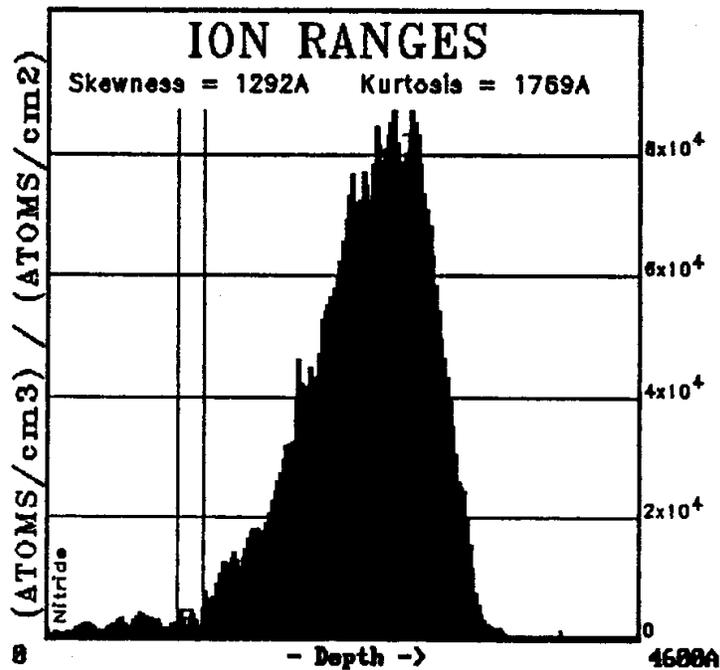


Figure 3: Range calculation.

much less since the nitride cap layer and the titanium glue layer reduce the energy of the incident ions. By the time the ions reach the deuterated palladium film their energy will be down to about 27 keV (two keV lost in the Ti and 11 keV in the nitride). As can be seen in Figure 4, the neutron dose from a one milliAmp beam is 1.1 mrem/hr at one meter. The actual dose should be much less in our experiment since a typical current is only about 30 microAmps.

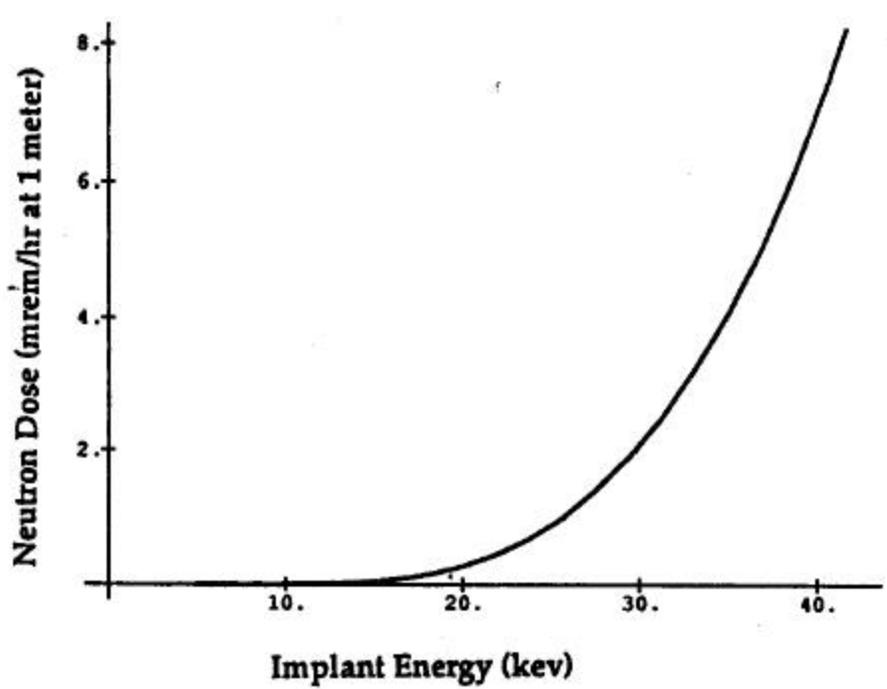


Figure 4: Calculated neutron dose from induced fusion.

THE ION IMPLANTER

Implantation was done with an Accelerator Incorporated ion implanter at IICO corporation in Santa Clara, CA. Deuterium was obtained from a heavy water source. During implant the 80 keV D_2^+ beam current varied between 10 and 45 microAmps. A solid source of LiF also was available to provide a 35 keV 6Li beam of 8 microAmps.

Ions from the source were preaccelerated, magnetically analyzed, accelerated, and then scanned across a two cm^2 circular aperture in order to produce a sharply defined spot on the target in the end station. The exact location of this spot was identified by securing a piece of paper to the target plate and exposing it to a low-current beam so that a burned area was created. The target plate was made of stainless steel and had a cooling loop soldered to its back surface. During implantation liquid nitrogen could be introduced into the cooling loop via a funnel accessible from outside the implanter end station.

FIRST EXPERIMENTAL RUN

Detector #9 was secured to the target plate as shown in Figure 5. In order to satisfy the conflicting requirements of good thermal conductivity but zero electrical conductivity between the device and the plate, a layer of thermal conducting material intended for use with power transistors was placed between the detector and the holder. A thin layer of vacuum grease was used to improve thermal contact between adjacent surfaces. Vacuum compatible Kapton tape secured the detector assembly in place. Liquid nitrogen was maintained in the target cooling

loop at all times during the experiment. We intended to monitor the temperature of the device by measuring the detector leakage current. However, it proved to be impossible to do this while implantation was in progress.

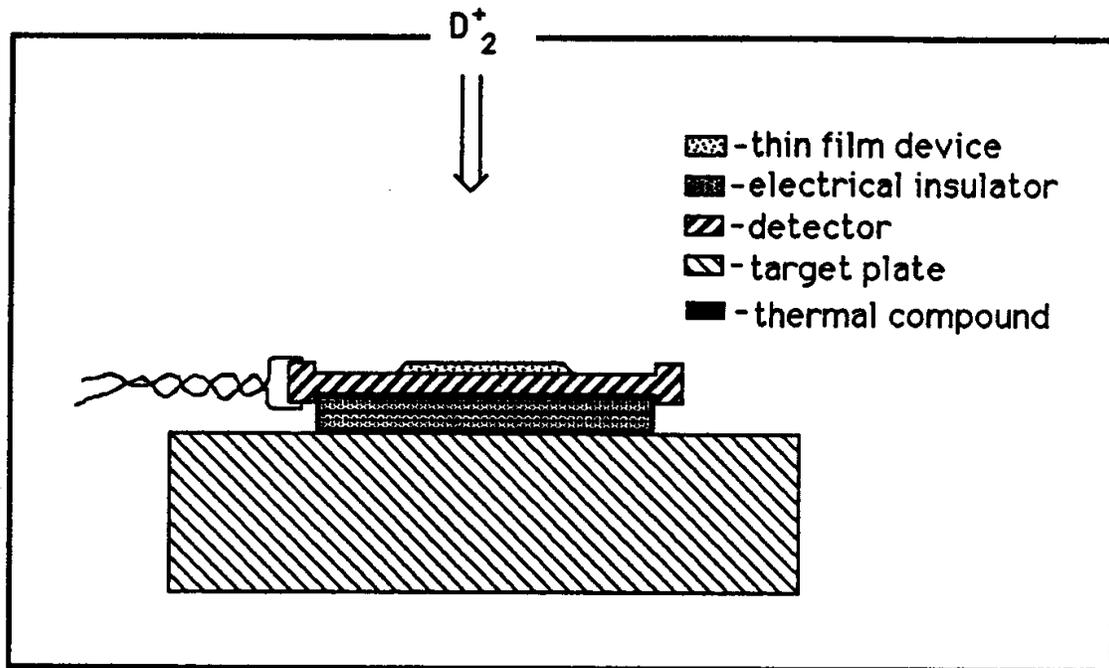


Figure 5: Detector #9 on target plate (first run).

A Tennelec TC908 supply set at -50 Volts was used with an Ortec 7402 preamplifier which had been modified to be compatible with the low resistance of the detector at room temperature. The output of an Ortec 575A amplifier was connected to a Davidson 4106 Multichannel Analyzer and to a Tektronix 2440 digital storage scope. An HP Think Jet printer was used to record the data from the scope. Excessive electrical noise made it impossible to look for pulses during implant.

A nine inch diameter Bonner sphere was available for neutron monitoring. This sphere was placed 40 cm from the detector outside the end station chamber as shown in Figure 6. Two Geiger counters were placed the same distance away.

Implant dose was monitored in the usual way with a charge integrator. The measured dose of course would equal the actual dose retained in the film only if the nitride capping layer survived or if the device could be kept cold. After a deuteron dose exceeding 100% had been implanted in detector #9, the implanter was shut down completely (except for the pumps) so that we could look for pulses from the detector. None were observed. A solid source of LiF, which provided ${}^6\text{Li}$, was installed in the implanter. A dose of 3×10^{16} ${}^6\text{Li}/\text{cm}^2$, equivalent to 1.3%, was implanted in the deuterated palladium film. After this, more deuterium implantation was done for a total nominal dose of about 150%.

After the above implants, the implanter again was shut down completely (except for the pumps) and signal monitoring was resumed. Apparent charged particle pulses were observed and recorded by the digital scope. The rate of these pulses initially exceeded 10 / minute. Typical pulses are shown in Figure 7 along with an ${}^{241}\text{Am}$ calibration pulse which had been taken prior to implant. Unfortunately, spectra of these apparent charged particle pulses could not be obtained due to malfunction of the MCA. During the same period in which the detector pulses were observed, both Geiger counters registered bursts of activity well above background. Breakdown of the high voltage cable to the neutron counter rendered its response unusable.

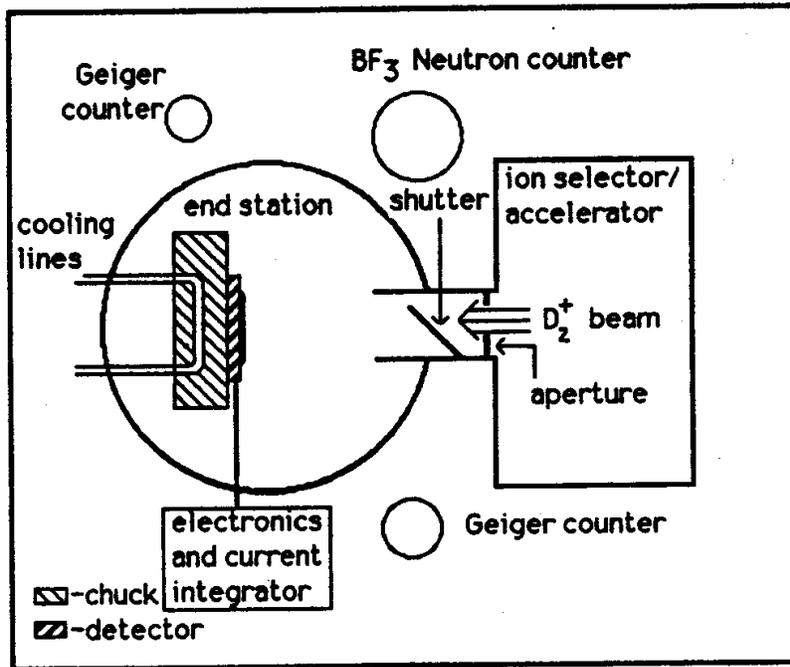


Figure 6: End station configuration for first run.

The apparent charged particle pulses and the Geiger counter activity persisted for four hours until it was necessary to warm up the device and remove it from the implanter so that the company could conduct its business.

A post-run spectrum showed no pulses from the detector due to possible stored charge effects.

THE SECOND EXPERIMENTAL RUN

Figure 8 shows the changes in the way the detector was installed on the target plate during the second run. This time a one mm thick copper disk was placed between detector #8 and the target plate in order to obtain better thermal contact. Electrical isolation was provided by a 1.5 mil thick sheet of Kapton. A stainless steel Faraday cup was placed on top of the plate to shield the detector from stray particles. A T-type thermocouple was epoxied to the back of the detector so that the temperature could be monitored continuously during implant. (The thermocouple wires had to be shorted together when the detector was being monitored in order to eliminate noise.) As shown in Figure 9, a ^{252}Cf source was attached to the shutter of the end station so that it could be swung into position to illuminate the detector and then swung back out of the way facing the implanter chamber wall. The alphas from this source are 82% with energy of 6.03 MeV and 15% with energy of 6.08 MeV. This provided an *in situ* calibration line.

The TC908 supply was replaced by a well-shielded battery box with a -67.5 Volt bias, and the Ortec 7402 was replaced with a low noise charge sensitive preamplifier from Lawrence Berkeley Laboratory. A shielded junction box was also set up on top of the end station cover.

An extremely sensitive neutron detector read out with a Ludlum 2200 Sealer Rate Meter was also used in addition to the nine-inch Bonner sphere. This sensitive neutron counter was placed about a meter from the thin film device as a monitor of the induced fusion rate as film loading progressed. It should be noted that fission neutrons from the ^{252}Cf source were well above the background rate.

Before the implantation began, detector 08 was tested for radioactive contaminants. An eight hour spectrum was taken with no peaks.

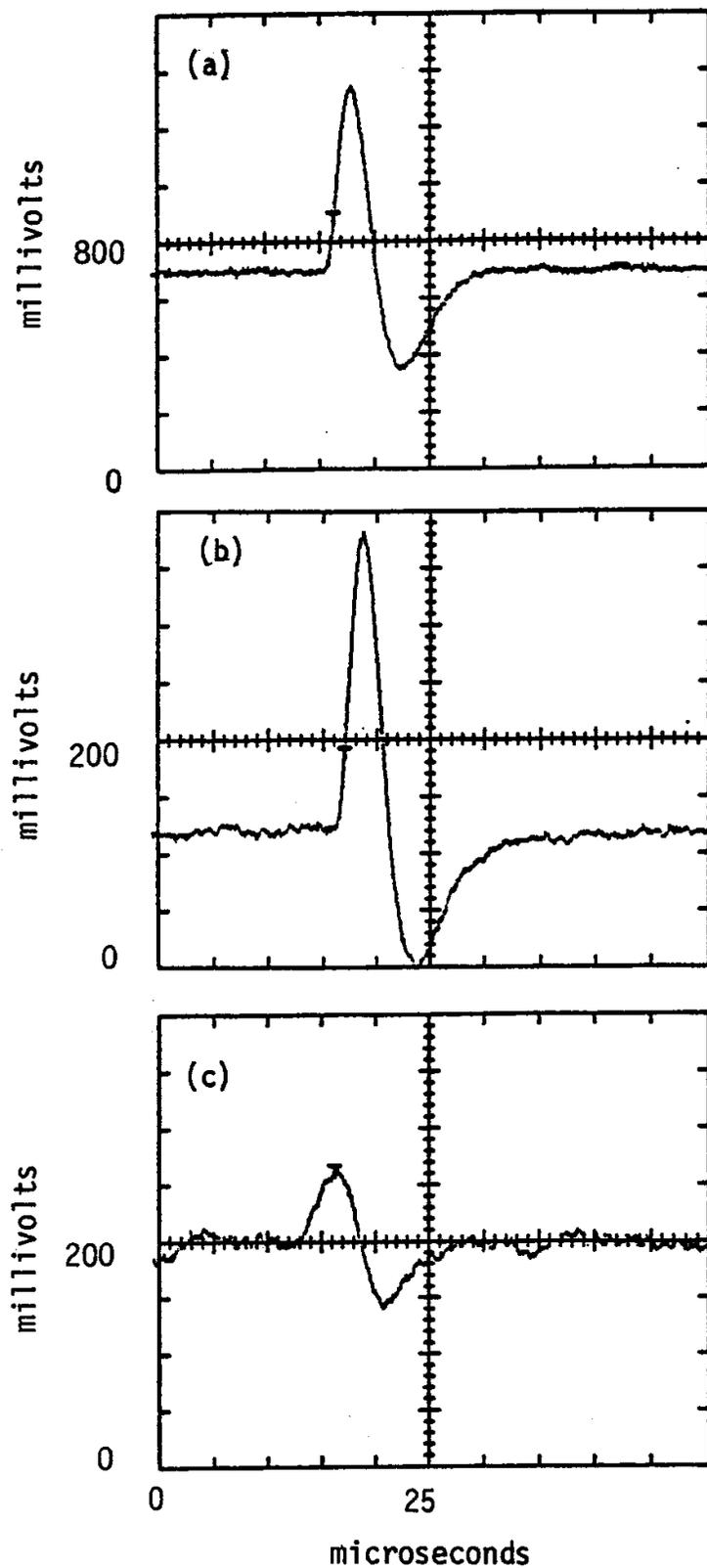


Figure 7: (a) ^{241}Am calibration pulse and (b), (c), typical apparent charged particle pulses observed at end of first run.

The detector then was implanted with the 80 keV D_2^+ beam. The temperature of the detector was maintained at all times, even during implant, at or below 200 K. Spectra were taken after 26%, 55%, 100%, and 150% deuterium loading, with the implanter shut down completely

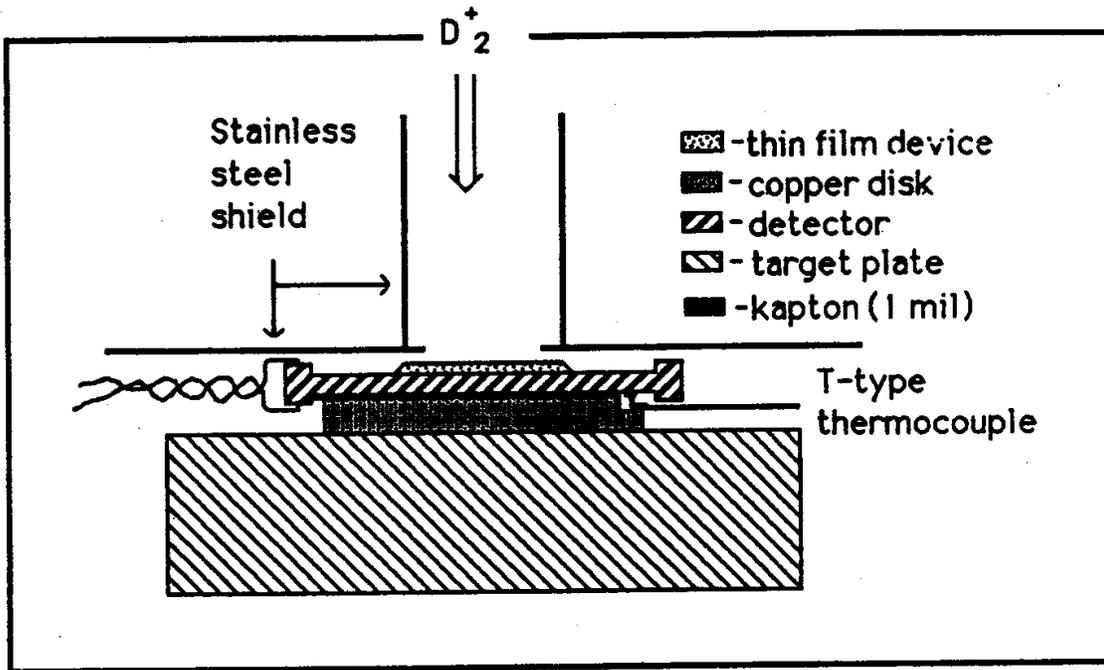


Figure 8: Detector #8 on target plate (second run).

(except for the pumps). One spectrum taken for 1000 seconds live time at nominal 100% loading is shown in Figure 10 with the ^{252}Cf calibration line present, and another spectrum is shown in Figure 11 at nominal 150% loading without the calibration source. No charged particle pulses were present. The device was allowed to warm to room temperature. Again a spectrum was taken with no pulses present.

Figure 12 shows the neutron count per microAmp as a function of implant time. There is an

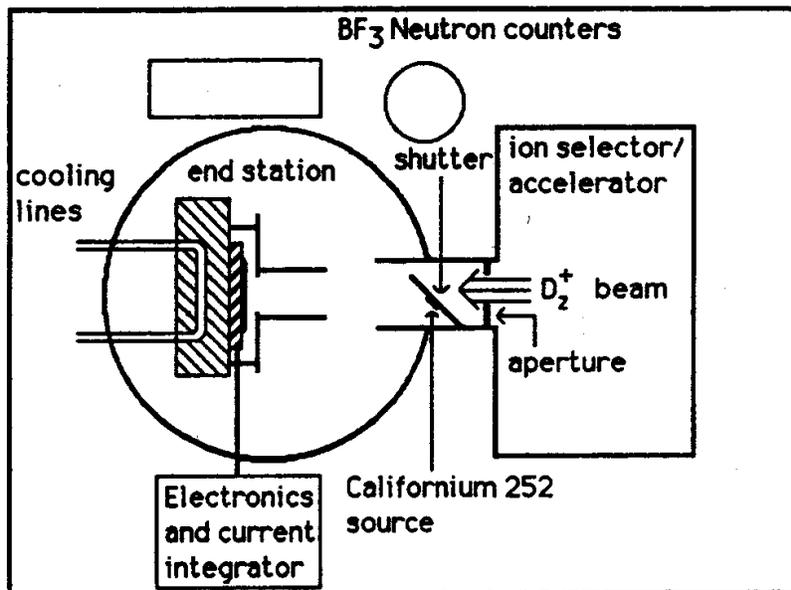


Figure 9: End station configuration for second run.

increase in neutron production up until 240 minutes of loading. After that the neutron production rate leveled off and even decreased at about 260 minutes. This is a strong indication that the

nitride cap failed catastrophically during the implant so that high deuterium doses were not achieved. Inspection of the device showed that the film had indeed cracked and flaked.

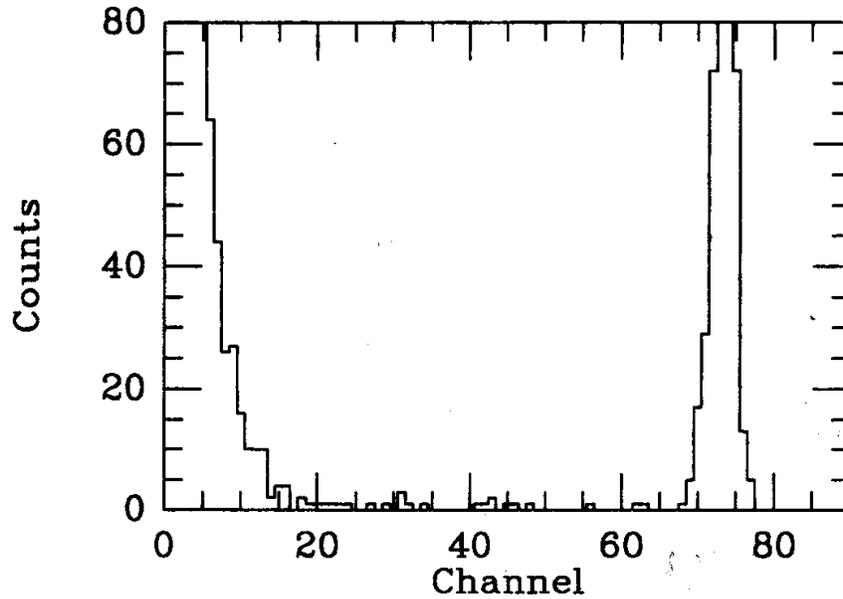


Figure 10: Spectrum with ^{252}Cf calibration line taken at nominal 100% loading during second run

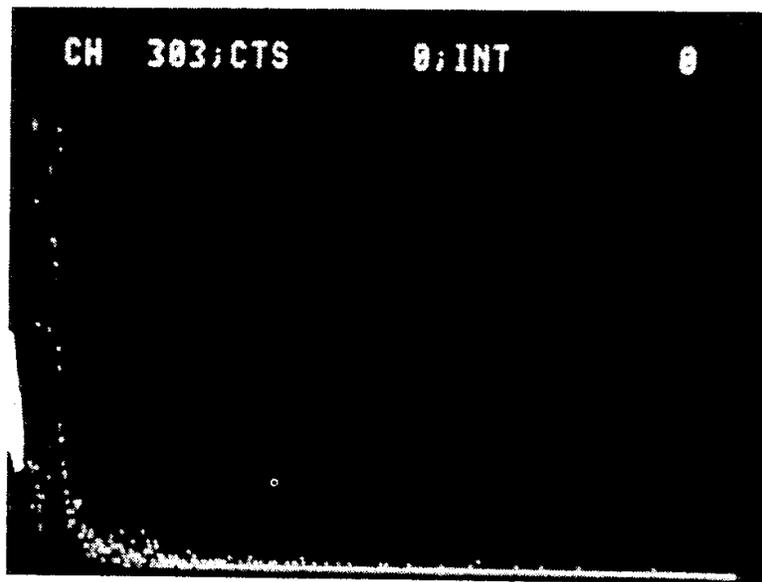


Figure 11: Spectrum taken without calibration line at nominal 150% loading during second run.

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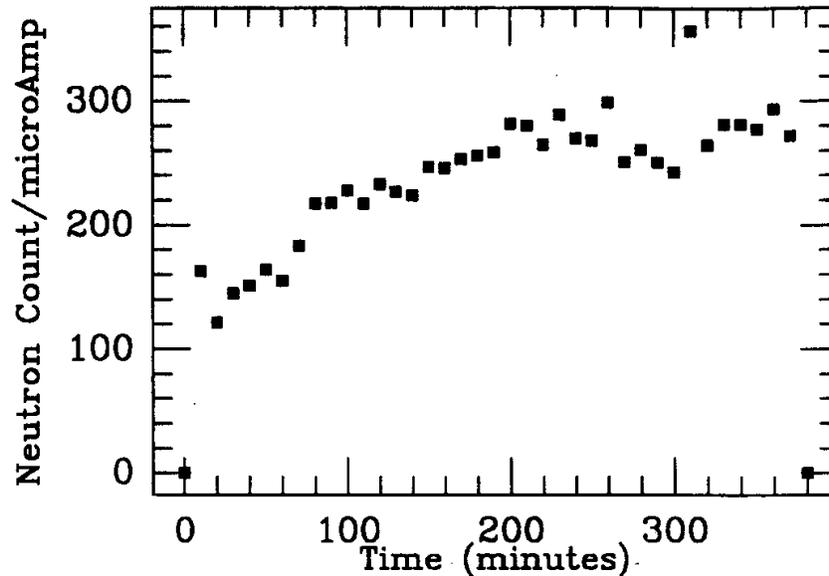


Figure 12: Normalized neutron production rate due to induced fusion during second run.

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