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PRELIMINARY RESULTS FROM THE BYU CHARGED-PARTICLE SPECTROMETER

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INTRODUCTION

In early papers, we noted the possible release of tritium from a volcanic source implying the occurrence of natural fusion [1,2]. At this conference, our collaborator Fraser Goff (Los Alamos National Laboratory) showed data pointing to tritium content of about three TU (Tritium Units) in magmatic water sampled from fumaroles of Mt. St. Helens.[3] Peter Britton (Reiss Foundation) measured increasing levels of both tritium and helium-3 with depth in boreholes in the Hamilton Shear Zone, an intriguing result.[4]

While the fascinating hypothesis of natural fusion in the planets [1,2] is being explored, we have begun complementary experiments to look for charged-particle production. Our detector is easily capable of detecting production of charged particles of MeV energies at the level of 0.06 particles per second, the average rate for neutron production given in Ref. 2. We note, however, that production at this rate over a few hours typical of the neutron data [2] results in production of only ~10^3 atoms in the laboratory. Nuclear products in atomic form at such levels could not be distinguished from normal contamination levels. The direct measurement of energetic charged particles emanating from thin films is far more sensitive than measurement of changes in tritium or helium or isotopic levels in samples. Moreover, the energy of any observed particles carries crucial information regarding the reactions which generated them.

We are further motivated in this study by the recent experiments of E. Cecil [5], R. Taniguchi [6], X. Z. Li [7], G. Chambers and G. Hubler [8] and others whose results show nuclear particle production at low but easily measurable rates in deuterided metal foils.

Here we review the detection system developed at BYU for charged-particle studies, thin foil target preparations, and observations of MeV-particle signals typically lasting for minutes to hours. These events appear inconsistent with radon decay or with cosmic-ray-induced events, but consistent in rates and episode-durations with observed neutron emissions.[2] The identity of the charged particles and the reactions which produced them are not yet established.
DETECTION SYSTEM

We have coupled a charged-particle counter and a neutron spectrometer as shown in Figure 1 (next page). The neutron spectrometer is similar to that described in Nature [2] except that solid plastic scintillator is used in place of liquid scintillator and only one photomultiplier tube is used. PMT pulses are digitized at 100 MHz using a LeCroy waveform digitizer over 160 μs and data are stored using an IBM-386 PC/CAMAC system. Thus, we can analyze pulses off-line and differentiate pulses from: noise (ragged shapes), cosmic-ray muons or gammas (plastic-only pulses, usually large), or neutrons (plastic-start pulse followed in ~11 μsec by a glass-capture pulse); see Fig. 2. The neutron spectrometer is also an effective cosmic-ray veto counter since most cosmic rays which strike the thin scintillators of the charged-particle counter must also pass through this counter; see Fig. 1. The neutron spectrometer is further discussed in Ref. 9.

The charged-particle spectrometer (CPS) incorporates a very thin plastic scintillator (0.0157 gm/cm$^2$) glued (0.0073 gm/cm$^2$ silicon glue) onto a thicker glass scintillator (0.375 gm/cm$^2$) which is glued onto the face of a 12.7-cm diameter photomultiplier tube (PMT). PMT pulses are again digitized at 100 MHz over 160 μs, synchronously with pulses from the neutron detector. That is, if a pulse is detected in either detector, we store digitized signals occurring 60 μs before and 100 μs after that pulse from both detectors. Pulse-shape analysis allows us to distinguish plastic pulses from glass pulses which are broader in time. The integrated area under the pulse reflects the light output of the scintillator which corresponds to the energy of the incident charged particle. As shown in Fig. 3, the plastic scintillator has a non-linear response in that the light output depends on particle energy and identity. The plastic scintillator thickness is chosen to just stop 5 MeV tritons and therefore to transmit 5 MeV protons; such protons would then deposit 2.7 MeV in the glass.
Figure 1.

Coupled Charged-Particle Detector / Neutron Spectrometer at BYU

Plastic scintillator
Glass scintillator
5" PM tube
Target Foil

Neutron Spectrometer
(See papers by Cecil [5] and Chambers [8] which provide striking evidence for production of tritons at such energies in partially deuterided foils.)

The CPS has the virtue of large surface area relative to surface-barrier detectors so that a relatively large sample can be studied in each experimental run. When used in tandem as shown in Fig. 1, the detectors are housed in a light-tight box with electrical feed-throughs. We have also run the charged particle detector by itself in a vacuum chamber we designed with electrical feed-throughs, to exclude radon.

DETECTOR CALIBRATIONS

The neutron spectrometer was calibrated using 2.45-MeV neutrons produced by the d-d reaction facility at the Colorado School of Mines, with the assistance of Prof. Ed Cecil. Fig. 4 (next page) displays the response of the spectrometer to 2.45-MeV neutrons, and shows that the calibration spectrum is indeed similar to that extracted from our original Nature papers [2,10] thus reinforcing the results presented there.

Figure 5 displays the response of the charged-particle spectrometer (CPS) to 5.45-MeV alpha particles from an americium-241 source. The source sits on a sheet of aluminized mylar, 1.4 μm thick (the usual “cover” for the photomultiplier tube).

Here we plot the pulse-shape versus the integrated pulse area and obtain a well-defined cluster on the two-dimensional plot. The pulse-area reflects the 5.45 MeV energy of the alphas, and the projected histogram of the cluster along the horizontal scale indicates the resolution of the counter for 5.45-MeV alphas.

![Figure 5. Response of the charged-particle spectrometer to 5.45-MeV alpha particles from an americium-241 calibration source. Dotted line indicates selection of plastic-like pulses: points above line are projected to horizontal axis to form histogram of particle-energies.](image-url)
Figure 4. Measured response of neutron spectrometer to 2.45 MeV neutrons from d-d reaction facility at Colorado School of Mines (b), juxtaposed with cold-fusion data (a) extracted from original Nature papers [2,10].
TARGETS

We began our investigations with Ti foils approximately 30 \( \mu m \) thick and partially hydrided with either H\(_2\) or D\(_2\) (ref. 12 gives treatments used). These foils had been prepared several weeks before use in this experiment then were cycled in liquid nitrogen before use as targets with the detector system. We saw no evidence of charged-particle production over seven weeks of running with these foils, but built up an understanding of our system, of cosmic-ray events, and beat down the background with shielding.

Motivated in part by the work of X.Z. Li [7], we next tried Pd foils 25 \( \mu m \) thick, annealed in vacuum at about 600\(^\circ\)C, then loaded with a mixture of 75\% D\(_2\) + 25\% H\(_2\) (or pure H\(_2\) for control runs) at 12 atm. After cooling, each target foil was unrolled (it had been rolled into a cylinder to fit into the stainless-steel sample cylinder for gas loading). The Pd foils were noticeably more brittle when deuterided and were crinkled upon unrolling. Each foil, approx. 1-3 in. diameter, was then placed on the face of the counter in the configuration shown in Fig. 1, on top of a thin (\( \sim 1.4 \mu m \)) sheet of aluminized mylar (to block any light flashes from the foil itself).

Our approach in cold-fusion experiments has been to produce non-equilibrium conditions in partially-deuterided solids [2,12]. In this experiment, non-static conditions result from deuterium, which had been loaded into the metal foil from the gas phase, diffusing through and out of the foil. We also induce temperature variations and concomitant stresses and phase changes by immersing a foil in liquid nitrogen for several minutes prior to some runs.

BACKGROUND, SOURCE STUDIES, RADON

Figure 6 shows the pulse-shape (plastic or glass scintillation) versus pulse-area (energy) distribution obtained with untreated 25-\( \mu m \) Pd foil. We endeavored to understand the origin of points in this distribution. By selecting events where the neutron counter shows a cosmic-ray-muon signal coincident (within 3 \( \mu s \)) with a signal in the charged-particle spectrometer (CPS), we select cosmic-ray events in the CPS and obtain Figure 6c. We see that cosmic-ray backgrounds account for much of the background signal in the glass scintillator. This is reasonable since the glass is sufficiently thick to register minimum-ionizing particles whereas the plastic is so thin that only low light output is generated. As seen in Fig. 6, the separation between plastic and glass pulses is pronounced, especially for light-outputs greater than that of the americium-calibration source (Fig. 5). Consequently, we can select plastic-like pulses (above the dashed line in Fig. 6b) and project these on the pulse-area (energy) axis, resulting in the histogram shown at the bottom of Fig. 6b. Figure 6e,f shows a typical pattern obtained with Pd foil treated with hydrogen as a control.
Figure 6. Pulse-shape (plastic or glass scintillation) versus area for Pd-foil (background) (a). Dotted line indicates selection of plastic-like pulses: points above line are projected to horizontal axis to form histogram of particle-energies (b). Selecting signals coincident with the cosmic-ray veto counter pulses gives distribution (c) whereas (d) represents points not vetoed. Figs. e, f correspond to (a,b) above and show patterns obtained with Pd-hydride foil for 7200 sec. Thus, 6e,f constitute Pd-H control run to compare with Pd-D foils (Figs. 10-12). (The pulse-shape measurement is the ratio of the area in the first 200 ns of a pulse vs the pulse’s total area.)
Figure 7 shows the patterns produced by cobalt-60 and californium-252 sources. Gammas from the Co-60 produce Compton-scattered electrons which register as low-light-output plastic and glass signals (Fig. 7a). Background gammas, for instance from decay of potassium-40 in concrete floors and walls in the BYU laboratory, will generate a similar pattern—as seen in the background plot Fig. 6a. The Cf source produces both gammas and neutrons, common low-level backgrounds in the laboratory even without sources nearby (the condition under which experiments were run). The pattern generated by the Cf source (Fig. 7b) is rather similar to that of the background. We note that energetic neutrons will generate some signals by producing recoil protons which register mainly in the plastic scintillator.

Radon is of particular concern in these studies since it produces alphas of energies up to about 10 MeV (Thorium-C' decay, Fig. 8a) and since it is present in laboratory air. We have gathered data relating to radon decay in Figure 8. Figure 8a includes an important branch in the decay of radon. Note that thorium-C (Bi-212) beta-decays to Th-C' (Po-212) which then decays via alpha emission with a half-life of 0.3 μs. Thanks to fast pulse digitization, we are able to clearly identify this decay sequence as shown in Fig. 8b, which displays a small beta-induced pulse followed by a relatively large and narrow pulse due to alpha-stop in the plastic scintillator. Figure 8c displays the pulse-shape versus pulse area plot obtained with a thin layer of thorium acetate powder (on 1.4 μm aluminized mylar) as a source. Selecting cases where two pulses occur in the CPS within 3 μs and the second pulse is plastic-like results in Figure 8d; in Fig. 8e the time cut is relaxed with little change. The characteristic time between the two pulses is shown in Figure 8f, where bin i gives the counts in all bins above bin i (thus integrating the exponential); the time spectrum is consistent with the expected half-life of thorium-C'. By comparison, Figure 8c'-8f' shows data treated in the same way but obtained with a Pd-hydride foil.

The similarity of the results shows clearly that radon was present on the foil. We have observed the same pattern and time distribution in some runs with Pd-H and Pd-D and untreated Pd foils. Consequently, many runs were conducted with the CPS in a vacuum chamber to exclude radon, although we found that some radon is held by the test foil for several hours following evacuation. The pattern of a small pulse followed within a few microseconds by a large plastic pulse (as in Fig. 8b) is characteristic of thorium-C' decay, an important branch of radon decay. It is one of the strengths of our detection scheme and pulse-digitization to be able to identify such decays; indeed, we are able to trace the presence of radon by registering the thorium-C' decay "fingerprint."

A major decay chain of radon involves uranium so we examined uranium acetate also. The pattern seen in Fig. 9 is reminiscent of that obtained with the Cf source (Fig. 7b) and does not differ greatly from the background
Figure 7. Patterns in the charged-particle spectrometer produced by (a) cobalt-60 and (b) californium-252 sources. Dashed-lines serve role described in Fig. 6.
Figure 8a. Thorium-series line of decay, showing how radon enters this chain. Adapted from Evans [11].
Figure 8b. Digitized pulses from the neutron spectrometer (upper line, one signal in d) and charged-particle spectrometer, lower dashed line. Note small pulse followed by larger plastic-like pulse [narrow in time], characteristic of thorium-C decay. Data obtained with Pd foils.
Figures 8c-f. Upper plots (c-f) show CPS patterns obtained using thorium-acetate powder as a source, evidencing beta decay followed by alpha production with a characteristic time of 0.3 us. A similar pattern was found with a Pd-H foil (c'-f') examined in air showing that radon was present in this case.
OBSERVATION OF CHARGED-PARTICLE EMISSIONS

Having described the detectors, targets, and backgrounds, we turn to observations taken with deuterium-charged thin Pd foils. In about three months of running, using both Pd + D_2 and Pd + H_2 foils plus frequent calibration runs to monitor small variations in electronics, we obtained several episodes of apparent charged-particle production from the deuterium-loaded foils only.

Let us scrutinize three cases (Figures 10-13).

Figure 10 shows data acquired in December 1990 with a Pd-deuteride foil placed in the detector system (see Fig. 1) immediately after removal from a cylinder bearing deuterium. Notice a cluster of points in the region of the plot representing plastic scintillations, and having a light output about 1.8 times that obtained with the Am-241 source (compare Fig. 5). Projecting points in the plastic-scintillation region in the 2-dimensional plot (i.e., above the dotted line) to the horizontal (energy) axis produces a histogram (Fig. 10b) with a striking peak at high energy where backgrounds are low (compare Fig. 7). Using the neutron counter in cosmic-ray rejection mode, we find that the peak remains (Fig. 10d): the cluster is not due to cosmic rays. We checked for the signature of radon decay via thorium: there are no events characteristic of thorium-C decay (see previous section) during the 7200-second run. Furthermore, the locus of data points in the cluster is lower in energy and narrower than one would expect from radon decay (compare Fig. 8d).
Figure 10. CPS spectra obtained with a 25 μm Pd foil impregnated with deuterium from the gas phase. Selecting plastic-like pulses and removing events with coincident cosmic rays gives the histogram in (d) showing a clear peak at high energy. Compare with background spectra, Fig. 6, which show no such peak.
Figure 11. (a) Rate of charged-particle production in the area of the high-energy peak shown in Fig. 10 for Pd-D, in ten-minute bins, as a function of run time. (b) Comparative rate of charged-particle production in the same energy region for background run with Pd-H.
Figure 12. CPS spectra obtained with a thin Pd foil loaded with 75% D2 and 25% H2 and placed in the tandem detector array (Fig. 1). A cluster of events survives the cosmic-ray veto cut (d) and appears at somewhat lower energy than in Fig. 10, perhaps representing a particle source deeper in the foil. Emission rate for this cluster is shown in (e) in 10-minute bins.
Figure 11 shows the rate of charged-particle production in the peak region as a function of time. The rate increased early on then decreased steadily over the duration of the two-hour run. The average count rate was 130 counts/hour for the first two hours. The CPS covers approximately 2π (see Fig. 1) and is nearly 100% efficient for detecting nuclear charged particles which strike it. Thus, the source rate was about 260 counts/hr, or 0.08 counts/second. This rate is comparable to the average rate for neutron production reported in our original paper [2], namely 0.06 source neutrons/second.

We suppose that there exists an active spot in the metal where fusion occurs at very low rates until the necessary conditions relax. Thus, the likelihood of a particle-production episode should increase with volume of properly-treated metal, but the rate from a single episode we would not expect to scale with volume of partially-deuterided material.

The signal-to-noise ratio here is about 35 (see Fig. 11), better than achieved to date in the neutron detector. Indeed, in this neutron spectrometer, we would expect only (260 neutrons/hr x 0.01 efficiency) = about 3 neutrons/hr, which cannot be seen above background in the current environment. Of course, this assumes that neutrons and charged particles (protons, tritons, helium-3 nuclei) are produced at roughly the same rate [2]. In the future, we will move the detectors deep underground and use our helium-3-proportional counter which has excellent gamma-rejection and neutron-detection efficiency of 31% (see Ref. 12). Thus we hope to meet our objective of measuring charged particles and neutrons in the same episode of nuclear activity.

Another example of evident charged-particle production is displayed in Figure 12. An untreated Pd foil was first examined providing the background pattern of Fig. 6. Then the foil was loaded with 75% D2 + 25% H2 and immediately placed into the detector array (Fig. 1). The pattern shown in Fig. 11 resulted, with a cluster evident in the two-dimensional plot and the histogram of plastic-like signals projected onto the energy axis. The energy represented by these emissions is somewhat less than in Fig. 10 (light output ~ 1.5 times that of Alphas from Am-241). This suggests that the particles came from an active region deeper in the foil so that particle energies were degraded on transmission through various angles from the foil. The cluster survives a cosmic-ray veto cut (Fig. 12d). Production rate versus time in the run is shown in Fig. 12e.

In an effort to exclude effects due to radon contamination, we ran several Pd-deuteride foils in vacuum with the charged-particle spectrometer alone. The experiments were performed in a different location in the underground laboratory with some minor changes in electronics, yet the effect survived. Figure 13 displays results from one run in vacuum. Again, there is a distinct cluster of points indicative of charged particles emanating from the partially-deuterided foil (Fig. 13a). We recalibrated the detector in this configuration...
Figure 13. CPS spectra obtained with Pd+D+H in a vacuum in an effort to exclude radon effects. (No signals characteristic of radon→ thorium-C decay were seen during this run.) A distinct cluster of points appears with light-output about twice that produced by an Am-241 source, with rate versus run time in (c).
using Am-241, and determined the cluster to have about 1.9 times the light output of 5.45-MeV alphas. Plotting the rate of particle production versus run time for this run (Fig. 13c) shows the rate was high for about 1.5 hours which is again consistent with duration of neutron-production episodes. There were no beta-alpha signals indicative of thorium-C decay (from radon) seen during this 1.5-hour period.

Thus, we have detected charged-particle production well above background levels. Detector responses allow us to rule out cosmic-ray origins of these charged particles. We hesitate to summarize exclude radon as the source of these clusters, although these were observed in vacuum and with no thorium-C decay detected. (Figs. 10-13).

What is the energy/identity of these particles? Close examination of digitized pulses from these events shows them to be clean plastic scintillations with no glass component, so the particles stopped in the thin plastic scintillator. We compare the light output associated with the clusters with that of the Am-241 source and use the light output versus energy plot (Fig. 3) to assess the particle energy/identity. We find that the observed clusters are consistent with 2.3-3.0 MeV protons and thus could arise from the reaction $d + d \rightarrow p(3.0 \text{ MeV}) + t(1.0 \text{ MeV})$. However, we cannot exclude the possibility that the clusters represent $d$ or $t$ up to 4 MeV, or alphas up to 8 MeV (see Fig. 3). In the near future we will use a movable Al foil as a degrader between the source foil and the CPS (in vacuum) to distinguish hydrogen-isotopic nuclei from alphas. We are also planning a second CPS using 75-μm thick plastic scintillator to improve particle identification. In addition, foils have been shared with other researchers (Cecil, Wolf, Li, Hubler, Bergesen).

CONCLUSIONS

We have designed and built a charged-particle spectrometer incorporating a thin plastic scintillator glued to a sheet of glass scintillator affixed to a 12.5-cm diameter photomultiplier tube. A metal foil charged with D$_2$ (or H$_2$) is placed onto the detector. A neutron spectrometer placed opposite the foil doubles as a cosmic-ray veto counter. All signals are digitized at 100 MHz and stored for analysis. We have studied the response of the detector to various sources and to backgrounds. Several episodes of charged-particle detection are reported. Detector responses are inconsistent with cosmic-ray (and provisionally radon) origins of the particles. The observed peaks are consistent with 3-MeV protons but could represent higher energy $d$, $t$ or He ions. Further studies are aimed at particle identification and at concurrent detection of neutrons in a deep underground environment.

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