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Charged particle detection in the Pd/D system: CR-39 SSNTD vs. real-time measurement of charged particle stimulated Pd K shell X-rays

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ABSTRACT

There have been a number of efforts to measure charged particle emissions in the Pd/D system. In general, two approaches have been employed. One approach was to indirectly detect charged particles by measuring Pd K-shell X-rays that should be created as charged particles traverse through the Pd lattice. The other approach utilized CR-39, a solid state nuclear track detector (SSNTD). With these detectors, a charged particle creates an ionization trail in the plastic that, upon etching, leaves a symmetric pit. The size, depth of penetration, and shape of the pits provides information about the mass, charge, energy, and direction of motion of the particles. While experiments done using CR-39 solid state nuclear track detectors have shown the presence of these charged particles, X-ray measurements of the Pd K-shell X-rays have not. The most significant difference between the two measurement techniques is that CR-39 is a constantly integrating detector and the X-ray measurements are done in real time. In this communication, this apparent discrepancy between the two charged particle measurement techniques is examined using known alpha sources.

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1. Introduction

There have been a number of reports of charged particle detection in Pd/D systems using CR-39, a solid state nuclear track detector (SSNTD). Li et al. [1] were among the first to use CR-39 SSNTDs in experiments involving hydrogen/deuterium gas loading of palladium. In these experiments, palladium foil was in direct contact with the CR-39 detector and the temperature was cycled between room temperature and liquid nitrogen temperature. No tracks were observed for the hydrogen loading experiments. However, a large number of tracks in the CR-39 were obtained as a result of the deuterium gas loading experiments. The lack of tracks in the hydrogen gas experiments indicate that the observed pitting in the deuterium gas experiments was not due to chemical attack. Price et al. [2] conducted similar gas loading experiments as Li et al. [1], however they observed no tracks above background. Unlike Li et al., Price et al. had cleaned their Pd samples with aqua regia. When Li et al. [3] conducted deuterium gas loading experiments with Pd foils that had been cleaned with aqua regia, they too observed no tracks. Auger analysis of the foils showed that chlorine had penetrated inside the Pd to a depth of a few hundred angstroms. When chlorine gas was used to intentionally contaminate the Pd surface prior to conducting the deuterium gas loading experiment, no tracks were observed in the CR-39 detector. This was the first indication that surface treatments could suppress the nuclear effects occurring inside the Pd lattice.

Lipson et al. [4] and Roussetski [5] used CR-39 detectors to detect charged particles emitted from deuterated Au/Pd/PdO heterostructures. In these experiments, the heterostructures were electrochemically loaded with deuterium. Once loaded, the heterostructures were placed in contact with the CR-39 detectors and the temperature was cycled to induce desorption of deuterium. Lipson et al. [4] reported seeing tracks consistent with 2.5–3.0 MeV protons and 0.5–1.5 MeV tritons in the CR-39 detectors. Besides tracks consistent with DD reaction products, Roussetski [5] reported observing triple tracks in the detectors used in his experiments. These triple tracks are diagnostic of the carbon shattering reaction, $^{12}C(n,n')3\alpha$, typically caused by a $\geq$9.6 MeV neutron.

Oriani and Fisher [6] were the first to report on using CR-39 detectors in an electrolysis experiment. They placed the detectors above and below the anode so as to not impede uniform loading of the cathode with deuterium. During each run, control detectors were immersed in a bottled electrolyte solution. Track densities ranged between 59 and 541 tracks cm^{-2} for the control detectors and 156–3760 tracks cm^{-2} for detectors used in active cells. They concluded that the reactions responsible for the particles causing

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the tracks did not occur at the distant cathode but most likely occurred in the electrolyte very close to the plastic surface.

Lipson et al. [7,8] were the first to conduct *in situ* electrolysis experiments in which the Pd foil cathode was in direct contact with the CR-39 detector. Experiments were done in both heavy and light water. The observed tracks were concentrated in areas where the detector was in direct contact with the cathode indicating that the Pd foil was the source of the particles that caused the tracks. The distribution of tracks was inhomogeneous. This indicated that some sites in the Pd foil exhibit greater activity than others. Using Cu and Al spacers between the cathode and the detector and linear energy transfer (LET) curves, they were able to identify the particles as being 11–16 MeV alphas and ~1.7 MeV protons.

Mosier-Boss et al. [9–11] used CR-39 detectors in their Pd/D co-deposition experiments. The optical properties of the Pd/D co-deposition generated pits were consistent with those observed for tracks of a nuclear origin. Specifically the pits were dark and circular in shape and they exhibited bright centers when focusing the microscope optics deeper inside the pits. Track density was highest where the cathode had been in contact with the detector indicating that the source of the tracks was the cathode. The distribution of tracks along the cathode was inhomogeneous indicating that some Pd sites were more active than others. Control experiments showed that the tracks were not due to radioactive contamination of the cell components nor were they due to chemical or mechanical damage. Tracks were observed on both the front and the back surfaces of the detectors. The only particles that can traverse through 1 mm thick CR-39 detectors are ≥40 MeV alphas, ≥10 MeV protons, or neutrons. The size and shape of the tracks on the front side resembled those observed for alpha particles with energies between 1 and 2 MeV. Scanning of a CR-39 detector, used in a co-deposition experiment in which a 6 μm thick Mylar film separated the detector from the cathode, showed that the majority of the tracks had diameters between 0.3 and 4.3 μm [10]. Protons with energies >10 MeV would produce tracks with diameters in this size range [12]. Mosier-Boss et al. [13] also reported on seeing triple tracks in CR-39 detectors used in Pd/D co-deposition experiments. As discussed *vide supra*, such triple tracks are diagnostic of ≥9.6 MeV neutrons. It was also shown that the Pd/D co-deposition triple tracks were indistinguishable from DT neutron generated triple tracks [14].

Tanzella et al. [15] also conducted Pd/D co-deposition experiments using CR-39 detectors. They conducted both light and heavy water experiments as well as experiments in which 6 μm Mylar separated the cathode from the CR-39. In the experiments where the CR-39 was immersed in the electrolyte, a 60 μm thick polyethylene film separated the CR-39 from the cathode. In the heavy water experiments, they also saw pits in the CR-39 detectors that correlated with the placement of the cathode. Tracks were observed on both the front and back surfaces. Lipson and Roussetski [15] analyzed the CR-39 detectors using a sequential etching technique that they had developed, using alphas and protons of known energies, to differentiate charged particles and their energies. Using this process, they identified proton recoils due to 2.45 MeV neutrons, 3 MeV protons, 16 MeV alphas, and 12 MeV alphas in the CR-39 detectors used in heavy water experiments. No tracks above background were observed in the light water experiments. It should be noted that the sequential etching method has been used by other groups working with radioactive materials to identify and determine the energies of emitted charged particles [16–19].

Prior to the use of CR-39 to detect charged particles in these experiments, attempts were made to detect the Pd X-rays resulting from the refilling of the K shell electron orbits ionized by the passage of charged particles through the Pd lattice [20,21]. Both Bennington et al. [20] and Deakin et al. [21] used lithium drifted silicon, Si(Li), detectors to detect the X-ray emissions in real time. In these experiments, no X-rays above background were detected. The main difference between the two approaches to detect charged particles is that CR-39 is a constantly integrating detector while the measurement of the X-rays using a Si(Li) detector is done in real time. In this communication, the apparent discrepancy between the two approaches in detecting charged particles in the Pd/D system is examined.

2. Experimental

2.1. Real-time X-ray/gamma ray measurements

All X-ray/gamma ray measurements were made using a cryogenically-cooled 18% HPGe detector with a Be window (Ortec). The detector head and samples were placed in a Pb cave. To reduce the Pb fluorescence, the inside of the cave was lined with Sn foil. Cu foil, between the Sn foil and the sample, was used to eliminate the Sn fluorescence. Sample orientation inside the cave is discussed *vide infra*. Compared to a Si(Li) detector, the HPGe detector used in these experiments is more sensitive to 21.1 keV Pd K shell X-rays.

All manipulations of spectral data were done using GRAMS/A17 (ThermoGalactic). This software package is used to subtract spectra interactively as well as integrate peak areas and measure peak intensities.

2.2. Silicon barrier detector measurements

Experiments were conducted stacking layers of 6 μm thick Mylar sheets on top of one another between the CR-39 detector and an ^{241}Am source. To determine the energies of the alpha particles getting through the Mylar, a silicon barrier detector (AMETEK model TR-SNA-300-100) was used. A ~0.5 mm wide slit was placed between the detector and the ^{241}Am source to block particles emitted at oblique angles. This allows particles that are approximately perpendicular to the plane of the Si barrier detector to reach the detector thereby reducing the backscatter. The slit used in these experiments was made from 100 μm thick acrylic plastic.

2.3. Etching of CR-39 and analysis of the etched detectors

The CR-39 detector was exposed to a ^{210}Po source for 3 min. Afterwards the detector was etched in an aqueous 6.5 N sodium hydroxide solution at 65–72 °C for 6 h. After etching, the detector was rinsed in water, vinegar, and again in water. Microscopic examination of the etched CR-39 detector was done using an Eclipse E600 epifluorescent microscope (Nikon) and CoolSnap HQ CCD camera (Photometrics). A magnification of 1000× was used. The software used to obtain the images was MetaVue (MDS Analytical Technologies).

3. Results and discussion

3.1. Overview of the 1989 X-ray measurements using Si(Li) detectors

The cell used by Deakin et al. [21] was constructed of Pyrex glass with a thin blown Pyrex window. The Pd foil cathode was 50 μm thick and had an area of 1 cm^2. The cathode was pressed against the thin Pyrex window. The X-ray detector was placed on the other side of the Pyrex window. The ability of the detector to register K shell X-rays from the Pd cathode was checked by fluorescing the electrode using Ba X-rays after the cell had been filled with electrolyte. They observed that room background radiation caused the Pd cathode to fluoresce and a line due to Pd K shell X-rays was present as an artifact in the background. After 333 h of electrolysis