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Reply to comment on "The use of CR-39 in Pd/D co-deposition experiments": a response to Kowalski

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Abstract. Earlier we reported, in this journal, that the pits generated in CR-39 detectors during Pd/D co-deposition experiments are consistent with those observed for pits that are of a nuclear origin. Recently, that interpretation has been challenged. In this communication, additional experimental data and further analysis of our earlier results are provided that support our original conclusions.

PACS. 29.30.Ep Charged-particle spectroscopy – 29.40.Wk Solid-state detectors

1 Introduction

CR-39 is a passive solid state nuclear track detector that has been used extensively in inertial-confinement-fusion (ICF) research [1]. The advantages of CR-39 for this application are that it is inexpensive, robust, it interacts with both charged particles and neutrons, and is insensitive to gamma and beta radiation as well as electromagnetic noise [1,2]. When a charged particle traverses inside CR-39, it leaves behind an ionization trail that is more sensitive to chemical etching than the rest of the bulk. Upon treatment with a chemical etching agent, tracks remain as holes or pits and their size and shape can be measured. The resultant tracks are conical in shape [3]. When the detector is backlit, microscopic examination of the surface of a CR-39 exposed to an alpha source shows that the tracks are elliptical or spherical in shape and are dark in color. When the microscope optics are focused on the bottom of the track, the tip of the cone of the track acts as a lens and appears as a bright spot. These are features diagnostic of a nuclear generated track.

Many of the attributes that make CR-39 attractive for use in ICF experiments also make it attractive for use in Pd/D experiments. Earlier a series of experiments involving the use of CR-39 detectors in Pd/D co-deposition experiments were discussed [4]. In these experiments, it was shown that pits in the CR-39 were obtained when Pd/D co-deposition was done on Ag, Au, or Pt wires in both the presence and absence of external magnetic or electric fields. When using a Ni screen cathode for the Pd/D co-deposition, pits were only obtained in the presence of either an external electric or magnetic field. The pits obtained as the result of Pd/D co-deposition are either circular or elliptical in shape, Figure 1a. When focused on the surface of the detector, the pits are dark in color. Focusing deeper inside the Pd/D co-deposition pits reveals bright spots attributable to the tips of the track cones, Figure 1b. As discussed above, these features are consistent with what is observed for pits obtained when the CR-39 detectors are exposed to alpha sources such as ^{241}Am or depleted uranium. In contrast, features that are due to background and chemical damage are bright, shallow, and irregular in shape. As discussed in the original paper, control experiments showed that the pits obtained in the CR-39 during Pd/D co-deposition were not due to either mechanical or chemical damage. The results of these control experiments are summarized in Table 1. No pits were observed when CR-39 was placed in contact with the cell components and plating solution. This indicates that the pits were not the result of radioactive contamination of the cell, cathodes, polyethylene support structures, heat shrink, or plating solution. Experiments were conducted in the absence of $PdCl_2$. No pits were observed indicating that the pits are not due to the impingement of D_2 gases on the surface of the CR-39. Experiments were conducted in which the $PdCl_2$ was replaced with $CuCl_2$. For both the $PdCl_2$ and $CuCl_2$ systems, oxygen and chlorine gas evolution occurs at the anode and a metal plates out in the presence of deuterium gas at the cathode, While metallic palladium absorbs deuterium atoms, copper does not. Pits were observed in the CR-39 used in the palladium electrodeposition but not the copper. These experiments indicate that the pits are not due to chemical reaction of the CR-39 with either D_2, O_2, or Cl_2 gases. Furthermore,

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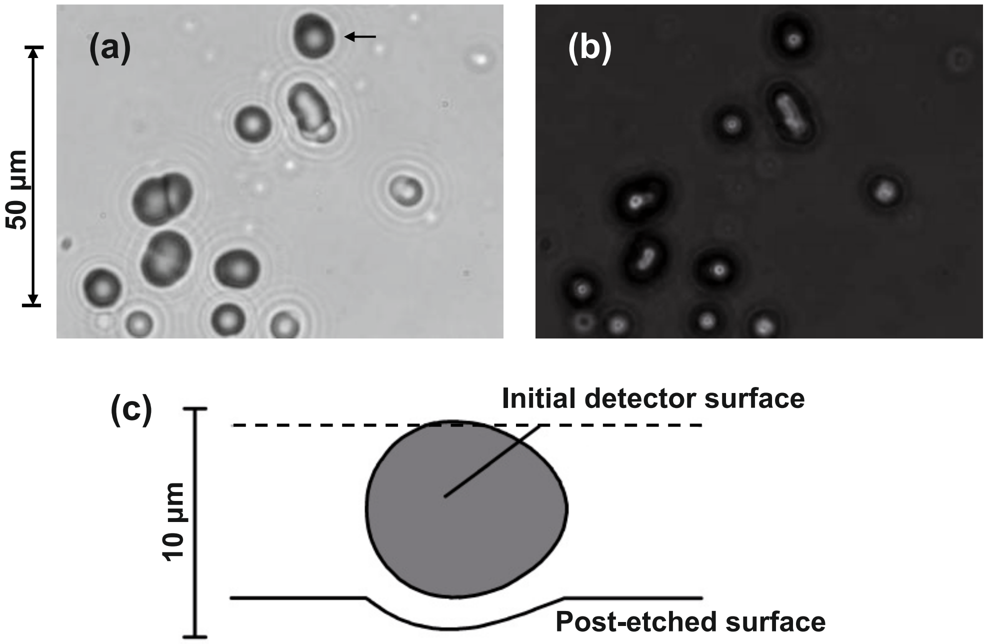

Fig. 1. Images of pits in CR-39 created as the result of a Pd/D co-deposition reaction. Ag wire, no external field experiment, 1000 × magnification. (a) Focus on the surface of the CR-39, (b) overlay of two images taken at two different focal lengths (surface and the bottom of the pits), (c) results of computer modeling the track indicated by an arrow in (a). This is the shape of the track obtained for a 1.3 MeV alpha particle hitting the CR-39 detector at a 35° angle after 6 h of etching at an etch rate of 1.25 μm h^{-1}.

Table 1. Summary of control experiments.

Experiment	Pits?	Conclusions
$PdCl_2$–LiCl co-deposition in D_2O	YES	Pits are observed where Pd deposit was in contact with CR-39. The Pd deposit is the source of the pits.
Cathodes, plating solution, $PdCl_2$ in contact with CR-39 – No electrolysis	NO	Pits are not due to radioactive contamination of the cell components
LiCl electrolysis in D_2O	NO	D_2 gas impinging on the surface is not responsible for the pits
$CuCl_2$–LiCl electroplating in D_2O	NO	Electrochemically generated D_2, O_2, and Cl_2 gases do not cause pits. Metal dendrites piercing into CR-39 not responsible for the pits.
$PdCl_2$–LiCl co-deposition in H_2O	YES	More than four orders of magnitude fewer pits are observed than for D_2O. Observed pits could be due to Pd/D interactions.

since both the Pd and Cu deposits are dendritic, the pits observed in the Pd system are not the result of dendrites piercing into the CR-39 detectors. In yet another series of experiments, D_2O was replaced with H_2O. Pits were obtained in the light water system but the density of tracks was at least four orders of magnitude less than was observed in the heavy water system. Since the natural abundance of deuterium in light water is 0.015%, it is possible that the tracks observed in the light water experiments could actually be due to Pd/D interactions.

The focus of the earlier paper [4] was to show that pits were obtained in the Pd/D system, to demonstrate that the characteristics of the pits were consistent with those observed for CR-39 exposed to energetic particle sources, and that the pits did not have a chemical or mechanical origin. There was no discussion as to the identity or energy of the energetic particles causing the pits as these aspects were and are still under investigation. However, Kowalski has recently written a paper commenting on the nuclear origin of the pits obtained as a result of Pd/D co-deposition [5]. Specifically he claims that the observed pits are too large to be attributable to alpha particles. In this communication we address the issues brought up by Kowalski. Specifically we discuss the results of spacer experiments using 6 μm Mylar between the cathode and the CR-39 detector as well as the results of track modeling.

2 Results and discussion

2.1 Energy of the particles produced as a result of Pd/D co-deposition

Kowalski [5] has compared the mean relative size of the pits obtained as the result of Pd/D co-deposition, Figure 5 [4], with those obtained after exposing the CR-39 detector to an ^{241}Am source, Figure 4 [4]. He measured the widths of the tracks and concluded that the Pd/D generated pits are 1.70 ± 0.17 times larger than the pits resulting from exposure to ^{241}Am. He then compared the ratio of 1 MeV alpha tracks with 4 MeV tracks using a calibration curve that was generated by Brun et al. [6] The ratio was 1.23 ± 0.12. Based upon this ratio, he claims that the Pd/D co-deposition pits are too big to be attributed to alpha particles.

There are a number of problems with the analysis done by Kowalski. For reasons that will be discussed *vide infra*, we have experimental data that suggests that the energy of the particles generated as a result of Pd/D co-deposition are on the order of 1 MeV. The energy of the alphas emitted by ^{241}Am is 5.6 MeV and not 4 MeV. Using Kowalski's calibration curve [5], the 1 MeV to 5.6 MeV ratio is 1.30 ± 0.13. In addition, the calibration curve that Kowalski used was generated using CR-39 obtained from Track Analysis Systems Ltd, UK and Pershore Moulding Ltd, UK. The CR-39 used in the Pd/D co-deposition experiments were obtained from Fukuvi. It is well known that there are variations in the quality of CR-39 between different manufacturers as well as batch-to-batch variations. To address this issue, we routinely expose one corner of our CR-39 detectors to an ^{241}Am source and use its alpha tracks as an internal standard. Consequently the calibration curve used by Kowalski is not applicable for Fukuvi CR-39. Using a calibration curve that was generated using Fukuvi CR-39 [7], the 1 MeV to 5.6 MeV ratio is 1.50 ± 0.15. Within the uncertainties, this ratio is in agreement with the 1.70 ± 0.17 ratio Kowalski reports for the comparison between the Pd/D generated pits and the ^{241}Am pits.

Experiments have been conducted using 6 μm Mylar between the detector and the cathode. In these experiments, a $\sim$90% decrease in the number of tracks is observed. Linear energy transfer (LET) curves indicate that the 6 μm Mylar cuts off $<$0.45 MeV protons and $<$1.05 MeV alphas. Consequently the majority of the particles are either $<$0.45 MeV protons and/or $<$1.05 MeV alphas. The energies of the particles are supported by computer modeling of the tracks [8,9]. The ionization trail created by the energetic particle traversing through the CR-39 is more sensitive to chemical etching than the rest of the bulk, i.e. $V_T > V_B$ where V_T and V_B refer to the rates of etching the track and the bulk respectively. For alpha particles, the relationship between V_T and V_B is described by the following equation [6]:

$$V_T = V_B \left(e^{(-a_1 x + a_4)} - e^{(-a_2 x + a_3)} + e^{a_3} - e^{a_4} + 1 \right) \quad (1)$$

where x is the residual range of the alpha particle and $a_1 = 0.1$, $a_2 = 1$, $a_3 = 1.27$, and $a_4 = 1$. Using the above equation, the track indicated by an arrow in Figure 1a was computer analyzed. This track has an elliptical shape indicating that the particle has entered the surface of the CR-39 detector at an angle. Computer modeling shows that a 1.3 MeV alpha particle hitting the surface of the CR-39 at a 35° angle will cause a track with a similar elliptical shape, Figure 1c. The ratio of the minor vs. major axis of the track indicated by an arrow in Figure 1a is 0.88. This matches the ratio (0.89) of the computer-generated track in Figure 1c.

2.2 Sequential etching

In addition to the control experiments to exclude chemical and mechanical explanations for the pits obtained as a result of Pd/D co-deposition, sequential etching of the CR-39 detectors was done [4]. These results were shown in the supplementary online materials. Figure S1[1] showed the changes in the pits obtained by exposure to an ^{241}Am alpha source as the detector was etched for 9, 12, 16, and 20 h. Figure S2 showed the changes observed for pits generated as the result of Pd/D co-deposition as the detector was etched for 9, 12, 16, and 20 h. As indicated *vide supra*, we expose one corner of the CR-39 detector to the ^{241}Am alpha source prior to running the Pd/D co-deposition experiment. The pits shown in Figures S1 and S2 were on the same chip. This assures that both kinds of pits were subjected to the same etching conditions. In our original paper we indicate that the pits seen in Figures S1 and S2 both exhibit the same kind of behavior as the detectors are etched, i.e. they get bigger, shallower, and more circular in shape the longer they are etched. Further discussion on the formation and growth of tracks as a function of etching time can be found in a review written by Nikezic and Yu [3].

In his analysis of Figures S1 and S2, Kowalski claims that the sizes of pits due to the alpha particles, Figure S1, keep growing between 16 and 20 h of etching while the Pd/D generated pits, Figure S2, remain the same [5]. Visual inspection of the pits in Figure S2 shows that the pits are getting larger [4]. Figures 2a and 2b show the 9 h etch time images obtained for the ^{241}Am and Pd/D co-deposition pits, respectively. Arrows in Figures 2a and 2b indicate the pits whose diameters were measured as a function of etch time. The ^{241}Am pit was clearly caused by an alpha particle that has impacted the CR-39 detector at

[1] Figures S1–S5 are only available in electronic form, section "online material" at http:/www.epjap.org/10.1051/epjap:2007152.

an oblique angle. The pit generated as a result of Pd/D co-deposition exhibits a circular shape. The 6 μm Mylar experiments suggest that the majority of the tracks generated have energies on the order of 1 MeV or less. Computer modeling, summarized in Figure 3, indicates that, for 1 MeV alphas, the angle of incidence cannot be determined by the shape of the pits. However, if the detector material is homogeneous, the track etch rate along the particle trajectory should be independent of the angle that the particle entered the detector [10]. Consequently, the ^{241}Am and Pd/D co-deposition pits can be compared directly. Figure 2c shows plots of pit diameter, as a function of etch time, obtained for the ^{241}Am pit, both major and minor axis, and the Pd/D co-deposition generated pit. As can be seen, the plots are linear and have similar slopes indicating that the ^{241}Am and Pd/D co-deposition pits are being etched at the same rate. This further supports the notion that the pits obtained during Pd/D deposition have a nuclear origin.

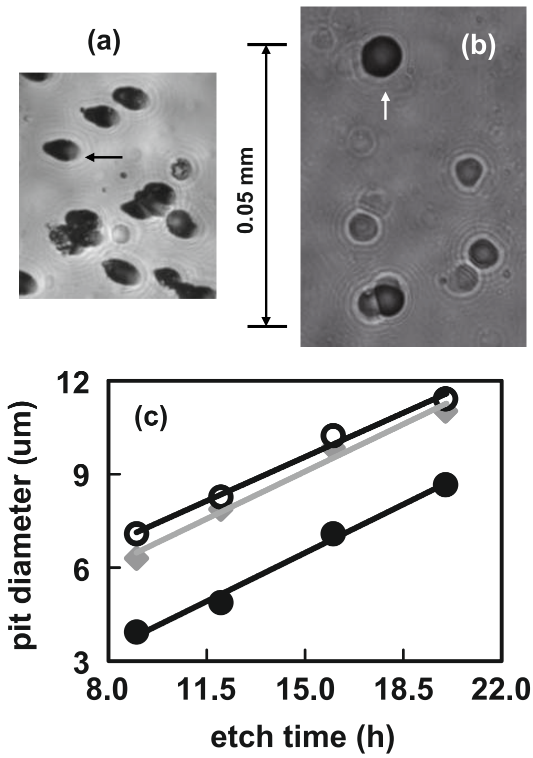

Fig. 2. (a) Image of pits in CR-39 created by exposure to an ^{241}Am source, 1000 × magnification, obtained after 9 h of etching. (b) Image of pits in CR-39 subjected to a Pd/D co-deposition experiment conducted on a Au wire in the presence of an external electric field, 1000 × magnification, obtained after 9 h of etching. (c) Plots of pit diameter (pits measured are indicated by arrows) as a function of etch time where: (○) corresponds to the Pd/D generated pit (the line has a slope of 0.404 ± 0.028); (♦) is the major axis of the ^{241}Am generated pit (the line has a slope of 0.434 ± 0.038); and (•) is the minor axis of the ^{241}Am generated pit (the line has a slope of 0.444 ± 0.028).

2.3 Corona discharge

Kowalski states that the pits generated during Pd/D co-deposition are as shallow as pits created in CR-39 by an induced corona discharge [5]. This statement implies that the pits are due to a corona discharge and do not have a nuclear origin. Again there are problems with this statement. As the modeling results indicate, Figure 1c, alpha particles with energies on the order of 1 MeV leave shallow tracks. Although Kowalski does not indicate this in his comments, the corona discharge experiment he refers to was conducted in air. It is nontrivial to create a corona discharge in an aqueous system. The use of corona discharge to degrade organic contaminants in water has been explored by others [11,12]. To degrade phenol in water, 15 kV were needed to create a corona discharge from a 50 μm diameter Pt tip electrode [11]. It was observed that as the solution conductivity increases, higher voltages were required to create the corona discharge [12]. The Pd/D co-deposition experiments are conducted in the presence of an electrolyte (either LiCl or KCl). Consequently the solutions are highly conductive. Also the measured cell voltages do not exceed 8 V. For these reasons, it is highly unlikely that we are creating corona discharges in our experiments. Finally, if the pits observed in Pd/D co-position were indeed due to a corona discharge, such pits should have been observed in the $CuCl_2$ experiments. As indicated in Table 1, no pits were observed in the $CuCl_2$ experiments. The $CuCl_2$ experiments also indicate that the pits observed in the Pd/D co-deposition cannot be attributed to triboelectrical charging of the CR-39 surface whether or not an additional dielectric layer (e.g. 6 μm Mylar) is present.

3 Conclusions

Earlier we reported that pits are formed on CR-39 detectors during Pd/D co-deposition. The pits are dark in color, circular/elliptical in shape, and have bright centers when focusing deeper in the plastic. These features are consistent with what is observed for nuclear generated tracks. A series of control experiments were conducted that indicated that the observed pits were not due to radioactive contamination of the cell components; or to impingement of the gas bubbles on the surface of the detector; or to chemical attack by D_2, O_2, or Cl_2 gases; or to the metal dendrites piercing into the plastic. More recently we have conducted experiments in which 6 μm Mylar is placed between the CR-39 detector and the cathode. Both the 6 μm Mylar experiments and computer modeling of the tracks suggest that the pits generated as a result of Pd/D co-deposition have energies on the order of 1 MeV or less.

Using a calibration curve for Fukuvi CR-39, it was shown that the ratio of 1 MeV to 5.6 MeV alphas is consistent with the ratio Kowalski reports for the comparison between the Pd/D generated pits and the ^{241}Am pits. Further analysis of the sequential etching results show that the Pd/D co-deposition pits and the 5.6 MeV alphas etch at the same rate. Because our cell voltages are between

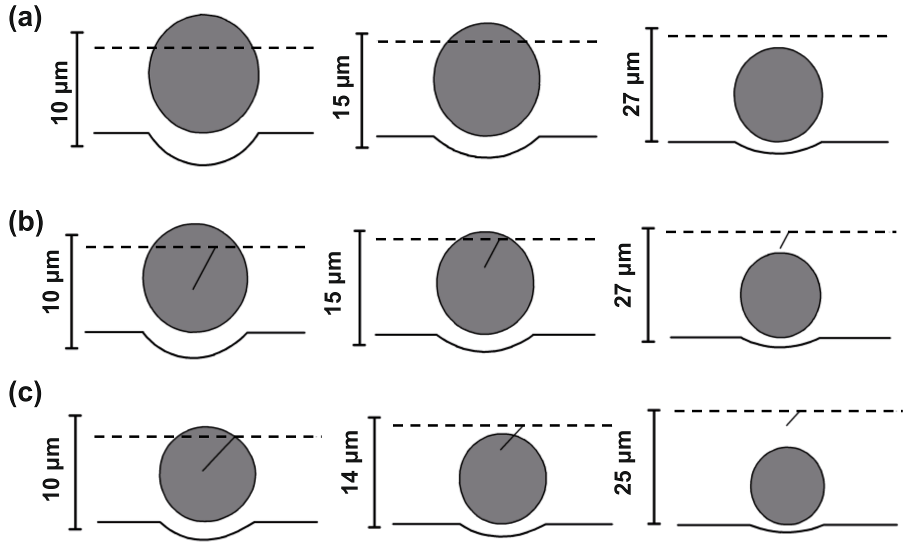

Fig. 3. Results of computer modeling 1 MeV alpha tracks after, left to right, 6, 10, and 20 h of etching. The etch rate is 1.25 μm h^{-1}. Equation (1) was used to calculate V_T. Angles of incidence used in the calculations are: (a) 90°, (b) 60° and (c) 45°. The dashed line above the track indicates the initial detector surface while the solid line below the track is the post-etched surface.

1–8 V and our solutions are highly conductive, it is highly unlikely that we are creating a corona discharge. Also no pits were observed in the $CuCl_2$ experiments. Therefore, the pits obtained in the Pd/D co-deposition experiments cannot be attributed to a corona discharge or to triboelectrical charging of the CR-39 surface.

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