Comment on "The use of CR-39 in Pd/D co-deposition experiments" by P.A. Mosier-Boss, S. Szpak, F.E. Gordon and L.P.G. Forsley

Interpreting SPAWAR-type dominant pits

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Abstract. A recent claim [Eur. Phys. J. Appl. Phys. **40**, 293 (2007)] demonstrating a nuclear process triggered by electrolysis is challenged. An analysis, based on relative diameters, is used to demonstrate that predominant pits could not possibly be attributed to alpha particles, or to less massive nuclear projectiles. This conclusion is supported not only by positive results from a replication experiment, but also by results from the experiment on which the original claim was based. While the numerous SPAWAR-type pits seem to be highly reproducible, their interpretation is not yet clear.

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1 Introduction

Co-deposition experiments described in [1] were performed by scientists from the US Navy's San Diego SPAWAR Systems Center (SPAWAR). The purpose of this note is to comment on some of these experiments. Are the predominant CR-39 pits, in SPAWAR-type experiments, due to nuclear particles created during electrolysis, as claimed by the authors, or are they due to something else? That is indeed an important question; the prevailing view is that chemical processes do not trigger nuclear processes.

As stated in [1], emission of charged particles during electrolysis has been reported as early as 2002 and 2003. Oriani et al. and Lipson et al., like SPAWAR researchers, used CR-39 detectors. But protocols developed by different teams of researchers were very different from each other. After learning about preliminary co-deposition results, the author of this note became one of several researchers who used the SPAWAR protocol and observed pits similar to those described in [1]. This was reported at the APS meeting [2]. Winthrop Williams from University of California, Berkeley [3], and the SPAWAR team [4] reported similar results. It became clear that experimental data are reproducible. That is important; results which are not reproducible belong to protoscience, not to science.

The authors of [1,4] claim that their "copious pits" are due to nuclear projectiles. Taking such a question-

able claim for granted, I will show that nuclear projectiles, if any, responsible for CR-39 pits, must be more massive than alpha particles. That was the main conclusion reached in [2]. Can the same conclusion be reached on the basis of SPAWAR's own experimental data [1]? What follows is an attempt to answer this question, and to comment on so-called PACA results. The acronym PACA, invented by Oriani, stands for Protected Against Chemical Attack. In the SPAWAR protocol CR-39 detectors are exposed to the cathode and to the electrolyte. In the PACA protocol [5], on the other hand, a thin mylar film (about 6 μ) protects the CR-39 detector from possible corrosive effects of the cathode and the electrolyte.

2 Relative sizes of predominant SPAWAR-type pits

Do SPAWAR experimental data [1] agree with the conclusion based on the replication experiment [2]? The answer is positive. SPAWAR predominant pits, on CR-39 chips in contact with the cathode during electrolysis, as illustrated in Figures S1 and S2, are also significantly larger than on chips exposed to alpha particles. The same conclusion can be drawn by comparing pits shown in SPAWAR Figures 4 and 5. Figure 4 shows about 30 pits due to alpha particles; Figure 5 shows 10 pits on a chip that was in contact

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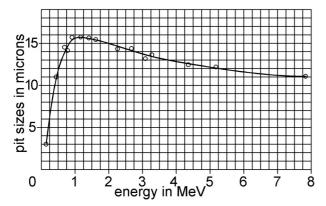


Fig. 1. Dresden calibration curve of CR-39 for alpha particles of different energies [7]. The etching solution was the 7.25 M NaOH at the temperature of 70 $^{\circ}$ C. The etching time was 7 h.

with the cathode during electrolysis. Microscopic magnifications are identical in these two figures. The mean width of pits in their Figure 5 is about 1.7 times larger than the mean width of pits in their Figure 4. Comparing widths, rather than lengths, is reasonable because lengths of pits often depend on angles of incidence.

The 1.7 ratio (plus or minus 10%) is significantly lower than the 2.5 ratio reported in [2]. Is that smaller ratio consistent with the idea that pits in Figure 5 can be attributed to alpha particles or protons? To answer this question, one should refer to the CR-39 calibration curve [6], shown in Figure 1 below. That curve refers to circular pits resulting from alpha particles intercepted at very small angles of incidence. Diameters of pits due to alpha particles of 4 MeV, for specified etching conditions, are 13 μ . This number would be different if etching conditions were different. The overall shape of the curve, however, would be essentially the same. Figure 1 can be used to predict diameters of pits due to alpha particles of different energies when a diameter is known for one particular energy, such as 4 MeV. It shows that even for the 1 MeV particles the expected diameters are not 1.7 times larger than diameters of pits due to 4 MeV particles.

According to that calibration curve, alpha particles of 1 MeV should produce pits whose diameters are only 16/13=1.23 times larger than those due to alpha particles of 4 MeV. The difference between 1.70 and 1.23 is 0.47; this is nearly three times larger than the estimated 10% uncertainly in the 1.7 ratio. In other words, the probability that mean diameters due to alpha particles of 1 MeV are 1.7 times larger than mean diameters due to alpha particles of 4 MeV is very small. The expected ratio for alpha particles of 2 MeV is 15/13=1.15. This is even more significantly different from the 1.7 ratio. Note that pits due to alpha particles of 7 MeV are expected to be about 15% smaller, not larger, than pits due to \sim 4 MeV particles, under identical etching conditions.

On the basis of these considerations, one can say that the ratio of widths, 1.7, based on SPAWAR data [1] is not consistent with the idea that their predominant pits are due to alpha particles. Their copious pits can also not be attributed to protons; pits due to protons are known to be about 30% smaller than pits due to alpha particles, at matching energies.

3 Consecutive etching: a powerful new method of investigation

A totally new approach to the problem of origin of postelectrolysis pits can be developed on the basis of Figures S1 and S2, presented in [1]. These figures show how pit sizes change when etching times become longer (9, 12, 16 and 20 h). It is remarkable that sizes of pits due to alpha particles (Fig. S1) keep growing between 16 and 20 h of etching, at a rate close to 2 μ per hour, while sizes of post-electrolysis pits (Fig. S2), remain nearly the same. This is another clear indication that post-electrolysis pits cannot possibly be due to alpha particles of 5 MeV. Note that, according to [7], pits created by nuclear projectiles are known to keep growing with etching time. For short etchings, profiles of pits are conical; for long etchings profiles become semi-spherical. A profile starts to be semispherical after the entire latent track is affected by the etching NaOH solution. Subsequently, diameters of pits grow at a rate depending on the temperature of the etching solution, and on its molarity.

Identification of pits due to nuclear projectiles, on the basis of consecutive etching, was first described by Russian scientists [8]. That approach seems to offer a powerful tool for either accepting or rejecting tentative explanations. Suppose that alpha particles of 1 MeV are suspected of causing pits on chips exposed to a cathode, during electrolysis. Such an hypothesis would be confirmed if alpha particles of 1 MeV, for example from an accelerator, were used in the same way in which 5 MeV particles were used in [1]. The hypothesis would be confirmed if the rates at which pits are growing were the same for post-electrolysis pits and for the pits due to alpha particles of 1 MeV; otherwise the hypothesis would have to be rejected. Tests based on sequential-etching are not limited to alpha particles. Suppose a researcher has a good reason for suspecting that post-electrolysis pits are due to carbon ions of 30 MeV (because pits due to such ions are expected to be larger than those due to alpha particles, after 9 h of etching). In such case carbon ions of postulated energy could be used to either validate or refute the idea. In general, an hypothesis, about particles responsible for unusual pits would be acceptable only if these pits grew at the same rate as pits due to postulated particles.

4 A new nuclear process or an artifact?

Detection of nuclear projectiles in the CR-39 polymer, as indicated in [1], is possible because such projectiles ionize and damage molecules. Latent tracks consist of

highly-localized regions of damaged material. The etching solution removes such material more rapidly than it removes the undamaged material. That is how latent tracks become microscopically visible, after etching. The authors of [9] were the first to publish the results showing that the high density pits, in SPAWAR-type experiments, are similar to those caused by an electrical effect. That conclusion was reached by showing that dominant post-electrolysis pits, created in another successful replication of a SPAWAR-type experiment [10], were about as shallow as pits created by a deliberately induced corona discharge. In both cases, the degree of localized damage was said to be less pronounced than in pits due to nuclear projectiles. Shallowness of pits was deduced from results of consecutive etching. The authors of [8] also noticed that many pits, on chip #2 from Williams' SPAWAR-type experiment [10], were too large to be attributed to alpha particles or protons.

5 PACA-type experimental results

A PACA-type experiment, with the same electrolyte as in a SPAWAR-type experiment, was performed by Tanzella et al. [11]. The CR-39 chip, used in their experiment #7, was surrounded by a thin (6 μ) mylar film. This was done to eliminate direct contact with the cathode, and with the electrolyte. Only pits smaller than those due to alpha particles were found after 15 days of electrolysis. These pits were positively identified as tracks due to protons with energies between 2 and 3 MeV. Note that 6 μ , or 0.83 mg/cm², is close to the mean range of alpha particles of 1.5 MeV in mylar. Tanzella's result alone seems to indicate that predominant pits cannot be attributed to alpha particles with energies larger than 1.5 MeV.

The total number of pits, on both sides of the mylar-protected detector, was about 200. This translates into a mean density of $\sim 100~\rm tr/cm^2$. The background density, on an unused chip, was only 6 tr/cm². On that basis the authors concluded that detectors were not irradiated by neutrons from some unaccounted-for source. Using their own calibration curve, they showed that protons were indeed produced during electrolysis. Note that the mean density of $100~\rm tr/cm^2$ is many orders of magnitude smaller than typical densities of predominant pits produced in unprotected SPAWAR-type detectors, near cathodes. Earlier PACA-type experiments [4] also revealed presence of pits that, according to their sizes, were most likely due to protons, or alpha particles.

Assuming that protons resulted from elastic scattering of neutrons on hydrogen, the authors of [11] concluded that "presented experimental evidence can be considered as strong, unambiguous proof that the #7 detector was exposed to fast neutrons (2.5 MeV)". Knowing the track density, and assuming that neutrons were emitted isotropically, during the entire duration of electrolysis (15 days), they estimated the mean neutron emission rate. It turned out to be 3240 (plus or minus 500) neutrons per hour. Will

such interesting result be as reproducible as dominant pits are in unprotected CR-39 detectors? This remains to be seen. Will emission of 2.5 MeV neutrons during electrolysis be confirmed by using other kinds of detectors? This also remains to be seen.

The author of this note also performed several PACAtype experiments, as reported in [12]. Clusters of tracks due to nuclear projectiles were observed on several occasions. The results, however, were not reproducible.

6 Conclusions

Dominant pits in SPAWAR-type experiments are reproducible but additional evidence is needed to identify nuclear particles, if any, on post-electrolysis chips. Comparing sizes of dominant post-electrolysis pits with sizes of pits due to alpha particles shows that neither protons nor alpha particles can be responsible for dominant post-electrolysis pits. This conclusion, also reached in [8], is reinforced by results of SPAWAR consecutive etching [1]. Copious pits produced in SPAWAR-type experiments are not observed in PACA-type experiments. Pits discovered on chips used in PACA-type experiments could be due to emission of alpha particles or protons.

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