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Generation of Nuclear Tracks during Electrolysis

R. A. ORIANI[1]* and J. C. FISHER[2]

[1]*112 Amundson Hall, University of Minnesota, Minneapolis, MN 55455, U.S.A.*
[2]*600 Arbol Verde, Carpinteria, CA 93013, U.S.A.*

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We show that energetic charged particles are produced during electrolysis of a D_2O solution of Li_2SO_4 in a cell with a platinum anode and a palladium cathode. CR-39 plastic detectors, designed for recording alpha particles from radon decay, were immersed in the electrolyte during electrolysis. They recorded significantly larger numbers of energetic particle tracks than were recorded by control detectors not subject to electrolysis. Statistical analysis shows only a 3×10^{-6} probability that the electrolysis tracks and the control tracks could have arisen from a common population. We conclude that there is a causal relationship between electrolysis and the production of energetic charged particles. Because track formation requires particle energies substantially greater than thermal or electrochemical energies it seems inescapable that a nuclear reaction was responsible. [DOI: 10.1143/JJAP.41.6180]

KEYWORDS: electrolysis, heavy water, lithium sulfate, palladium cathode, CR-39 detector, particle tracks, energetic particles, nuclear reaction

1. Introduction

Nuclear reactions have been suggested as responsible for various anomalous observations in electrolysis experiments. Beginning with Fleischmann and Pons[1)] and followed by a number of others,[2–5)] there have been reports of heat in excess of that attributable to chemical reactions, to resistance, or to other known losses in the electrolyte. In most of these experiments the electrolyte was a solution of a lithium salt in heavy water (D_2O), and the cathode was palladium metal into which a significant concentration of deuterium was introduced by the electrolysis process. The anomalous heat was attributed to a new form of D-D fusion inside the cathode material. Searches for reaction products have been undertaken and reports have been published[6,7)] of ^{4}He in quantities consistent with generation in the reaction $D + D \rightarrow {}^4He$. Much smaller but significant amounts of tritium have also been reported,[8)] but there is no evidence for comparable numbers of neutrons.

Most theoretical treatments envision lattice-mediated enhancement of the rate of $D + D \rightarrow {}^4He$ fusion within crystalline materials.[9,10)] A minority envisions other nuclear reactions, an example being a chain reaction in the electrolyte involving massive neutron isotopes in interaction with lithium, producing ^{4}He as a reaction product.[11)]

If indeed nuclear reactions occur in the electrolyte, energetic charged particles might be made evident by means of CR-39 plastic detectors. When an energetic charged particle impinges on a CR-39 chip, or when an energetic neutron produces a charged recoil particle within or near the chip, the charged particle plows into the plastic material and leaves behind a persistent trail of internal damage. Appropriate etching causes the trail of damage to dissolve more rapidly than the surrounding undamaged plastic, and the resulting etch pit reveals the track. Early work has confirmed that the technique is capable of detecting energetic charged particles produced in association with electrolysis.[12)] The present work describes a series of experiments undertaken to clarify and quantify this phenomenon.

2. Experimental Procedure

The CR-39 plastic used in this research was BARYOTRAK obtained from Fukuvi Chemical Industry Company, Japan. It is pure CR-39 that retains its surface smoothness after etching. The plastic is received as chips about 15×5 mm in area and about 0.8 mm thick which are laser-inscribed with identifying numbers on one side. The chips are covered by plastic films in intimate contact with each side to prevent the formation of alpha tracks by radon in the air. The edges of the chips are unprotected. To prepare chips for our experiments they are halved by scratching on one side with a sharp knife and cracking apart the halves. The scratching is done through the protective film on the side of the chip that will not subsequently be examined for tracks. Very small holes are then drilled through the film-protected chips to enable their subsequent suspension by nickel wires in the etching solution. If necessary for identification, additional numbers or letters are inscribed on the chips by scratching with a sharp knife through the protective film.

Our experiments consist of determining the numbers of tracks that are produced during electrolysis. It is therefore necessary to count the tracks already in the chips prior to use. This is done by carefully removing the protective film and etching the chips by suspending them in 6.5N KOH solution at 60°C to 72°C for a period of 24 h to 6 h depending on the temperature. After rinsing off the alkali and drying, the inscribed sides of the chips are protected against radon by covering with adhering Scotch tape until photomicrography is carried out. Immediately prior to photography the chip is cleaned by scrubbing with a small brush under detergent solution, rinsing under tap water followed by distilled water, then drying in air. This procedure was found to be superior to ultrasonic cleaning. An area of approximately 0.08 cm^2 is photographed as a mosaic of ten or more adjacent images, each of which includes portions of the inscribed numerals for easy identification so that it can again be photographed after electrolysis is carried out.

It is necessary to distinguish between etch pits that identify charged particle tracks and those that result from

*E-mail address: orian001@tc.umn.edu

surface damage or internal defects in the plastic. Particle-caused pits are initially conical in shape with a sharp point that marks the deepest penetration of the etchant along the trajectory of track damage. Tracks enter the plastic at various angles to the surface, so that the perimeters of their etch pits can be circular or elliptical. Moreover, charged particles, from whatever reaction producing them, arrive at the plastic surfaces with residual energies that depend on their travel lengths in the electrolyte, and those with the least residual energies produce the shortest tracks. Etch pits from short tracks develop rounded tips, lose their conical shape early in the process, and have small perimeters. Such very small pits can be seen in plastic detectors exposed to alpha particles from pitchblende, in detectors exposed to radon in the air, and in the control and the active chips of the experiments. We accepted as representing true tracks conical etch pits with sharp tips, and also those pits with rounded tips where careful microscopic examination revealed a conical shape near the surface as the mean diameter decreased with increasing depth. We rejected etch pits that occurred in clusters and those that formed linear arrays fearing that they may have arisen from surface scratches. Identical criteria were applied to control chips and to active chips. Recognizing that there is an element of judgement in applying criteria for the acceptance of an etch pit as representing a true track, all measurements were made by only one of us (RAO) to achieve consistency.

A digital camera coupled to a computer and printer is used. Microscope magnification of 100× in bright field is employed for photography, although 500× magnification and dark field are also frequently employed to positively identify features as nuclear tracks. The output of the digital camera is processed by the computer by a ''Find Edges'' program to yield high-resolution, high-contrast prints. In all of these operations, great care is exercised to avoid touching with any hard bodies such as tweezers or glass those areas of the chip that will subsequently be examined, for experience has taught us that such contacts can produce unwanted surface damage.

The next step is electrolysis with detector chips suspended in the electrolyte. One or two chips supported by platinum wires are positioned above or below the anode. To serve as controls one or two pre-photographed chips are immersed in bottled electrolyte solution during the duration of the electrolysis. These chips have undergone the same series of operations as have the chips placed in the electrolysis apparatus. After the electrolysis is terminated both the control chips and the active chips from the electrolysis cell are cleaned, and their identified areas are again photographed.

The electrolysis apparatus consists of a cylindrical glass tube of about 15 mm internal diameter. Its upper end terminates in a taper joint to a fitting that contains a tungsten wire seal-through to which is spot-welded a platimum wire whose lower portion is a flat spiral that serves as an anode. The lower end of the glass tube terminates in a flanged joint between whose O-rings is clamped the cathode plate. The palladium cathodes are 25×25 mm sheets of 1 mm thickness obtained from Dr. E. Storms, Los Alamos National Laboratory (Retired). Pre-treatment of the cathodes consists only of sanding and washing. The electrolytic solutions of Li_2SO_4 in D_2O were of various concentrations near 0.025 g/cm^3. Electrolyses at current densities between 0.10 and 0.37 A/cm^2 were carried out for two or three days with the evolved gases released to the environment.

3. Experimental Results and Discussion

Following the first etch of an active chip, and prior to electrolysis, the nuclear tracks in each photographed area are noted. After electrolysis the second etch reveals new etch pits that arose during electrolysis. The new etch pits are counted and the counts are recorded. Dividing this number by the photographed area we obtain the track density for tracks formed during electrolysis. The same procedure was followed for the control chips.

The left hand column of Table I presents the results for chips that were exposed to electrolysis. The average density of new tracks for these active chips is 987 tracks/cm^2. The right hand column gives the results for control chips in which new tracks were presumably produced by radon in the laboratory environment during handling, etching, photography, and immersion in the D_2O–Li_2SO_4 solution over the same period of time as that for the electrolysis. On average the control chips recorded a new track density of 179 tracks/cm^2.

During electrolysis bubbles of deuterium gas may impinge upon the detector chips, but this does not happen in the control experiments described above. Therefore, on the off-chance that the bubbling might produce artifacts that could be confused with nuclear tracks, additional non-electrolysis tests were carried out in which deuterium gas bubbles, produced by forcing the gas through a fine fritted-glass tube inserted in Li_2SO_4/D_2O solution, impinged upon detector chips suspended in the solution by nickel wires. After etching they showed no evidence of artifacts.

The data in Table I strongly suggest that more tracks are produced in the active chips than in the controls. Most active chips have a larger track density than any control, and half of the controls have a smaller track density than any active chip. Yet there is a region of overlap in the distributions where active and control chips have comparable track densities. A careful statistical analysis is required to determine the significance of the difference between the two distributions.

We proceed as follows. The tabulated track densities are not symmetrically disposed about their mean values for the active chips, for the control chips, or for the combined sets of chips, indicating that these distributions are not gaussian. Because gaussian distributions are desirable for statistical analysis we work with the logarithms of the Table entries. The log(track density) values provide distributions that are close to gaussian as desired. We seek the probability that the two log(track density) distributions, one for control chips and the other for active chips, could have arisen by chance from a common population. The smaller this probability, the greater is the likelihood that the distributions are distinct and independent, and that there is a causal relationship between electrolysis and the production of etch pits. The mean value of the log (track density) for the active chips is 2.801 ± 0.098 where 0.098 is the standard deviation of the mean. The mean value for the control chips is 2.187 ± 0.060. The

difference between the mean values is

$$D = (2.801 \pm 0.098) - (2.187 \pm 0.060)$$
$$= 0.614 \pm 0.115 \quad (1)$$

where $\sigma = 0.115$ is the standard deviation of the difference. The ratio of the difference to its standard deviation is Students's t-value,

$$t = D/\sigma = 0.614/0.115 = 5.34. \quad (2)$$

For 33 degrees of freedom ($N - 2$ where $N = 35$ is the combined number of active and control chips) this t-value corresponds to a probability of 3×10^{-6} that so large a difference could have arisen by chance. We conclude that the active population is distinct from the control population and that the difference is caused by electrolysis. These results corroborate others[6,7,12] showing that nuclear reactions can accompany the cathodic deposition of deuterium upon palladium.

Comparison of photographic images of etch pits produced during electrolysis with those produced by alpha radiation from radon and from pitchblende shows that our results are consistent with the production of alpha particles, although we cannot distinguish between alphas and other charged particles that can produce similar effects because we did not investigate the etching rates and cone angles that might have enabled us to do so.

Among the control chips, fluctuations in the numbers of new tracks per image can arise from differences in radon exposure and in physical handling. Comparable fluctuations are expected to underlie the numbers of new tracks per image for the active chips but they cannot account for the wide variability of response that we observe, suggesting that one or more significant variables remained uncontrolled during electrolysis.

One uncontrolled variable is the positioning of chips in the electrolyte. By examining the active chips on the sides opposite to the inscribed sides on which etch pits were counted for the data in Table I, we found instances of large track density on one side with low track density on the other. As examples, in one experiment the inscribed side has a track density of 543 and the reverse side has 2120. In another experiment, the inscribed side has 2380 whereas the reverse side has only 300. We surmise that chips must be properly positioned in the electrolyte for optimal exposure to energetic charged particles from whatever nuclear reaction may be responsible. Intermittent activity of the nuclear reaction may be a second uncontrolled variable. We find instances of detector chips from the electrolysis experiments both of whose sides have very low track densities. This could indicate improper positioning of the detector chips or it could be evidence that the nuclear reaction did not occur in these experiments. Such intermittent behavior would be in keeping with the frequently observed irreproducibility of experiments aimed at detecting anomalous thermal energy generation or the production of helium or tritium. These two uncontrolled variables are probably responsible for much if not all of the excess variability among the active chips. Fortunately, in spite of the variability, statistical analysis was able to quantify the highly significant difference between the active and control populations.

Table I. Charged particle track densities in tracks/cm^2 for all active CR-39 chips exposed to electrolysis and for all control chips. (In a few experiments the number of active chips exceeded the number of controls.) They are ordered by magnitude with the control entries offset, to make evident that most of the active chips have larger track densities than any control and that half of the controls have smaller track densities than any active chip.

Active	Control
3760	
2756	
2375	
1733	
1138	
962	
962	
897	
757	
676	
578	
543	
318	541
315	260
	260
260	221
225	204
177	195
161	177
156	165
	143
	143
	118
	108
	95
	95
	75
	59

4. Conclusions

It is difficult to escape the conclusion that energetic charged particles, arising from an as-yet unidentified nuclear reaction, can be generated during the cathodic deposition of deuterium upon palladium. However, it remains unknown whether such particles can be generated by electrolysis upon other metals or by light water electrolysis. It also remains unknown whether the nuclear reaction responsible for charged particle tracks can be responsible for production of sensible thermal energy in excess of the normal production of energy in electrochemical processes. Although the present results are highly significant in demonstrating a nuclear reaction or reactions, the track density is many orders of magnitude smaller than would be expected for a reaction that produces the excess heat reported in other experiments.

Because of the short ranges of charged particles in the electrolyte, and because recoil particles are not expected, the reactions responsible for the particles that are detected by the CR-39 plastic probably did not occur at the distant cathode, but most likely occurred in the electrolyte very close to the plastic surface. It is likely that nuclear reaction was largely confined to electrolyte volumes that were not sampled by the

CR-39 chips, and that only exceptionally did turbulence from bubble agitation cause a wisp of reaction to approach a chip sufficiently closely to leave tracks. It also is possible that the vast majority of nuclear reaction products have too little energy to leave any tracks and that the ones we observed were caused by a rare side reaction.

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