J. Condensed Matter Nucl. Sci. 6 (2012) 108–117

Research Article

Nuclear Particles Generated by Electrolysis – a Review

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Abstract

CR39 plastic detectors for nuclear particles are employed to demonstrate that a nuclear reaction can accompany electrolysis, an electrochemical process. Detectors placed within or above the electrolyte display statistically greater numbers of nuclear pits than do detectors used as controls. Placing the cathodes in contact with Mylar-covered detectors is the basis for a technique that reproducibly generates evidence for a nuclear reaction during electrolysis. Nuclear pits are produced in plastic detectors exposed to the oxygen evolved at the anode, as well as to the hydrogen evolved at the cathode. Nuclear damage trails are found that are initiated within the 0.83 mm thickness of the plastic detectors. These phenomena clearly show that a nuclear reaction of as yet unknown nature can be generated during electrolysis.

Keywords: CR39 detectors, Damage trails, Electrolysis, Etch pits, Nuclear particles, Nuclear reactions

1. Introduction

In 1989 Fleischmann and Pons [1] presented calorimetric evidence that during electrolysis with lithium sulfate dissolved in heavy water and palladium as the cathode more thermal energy is produced than the electrical energy provided. Because the magnitude of the excess energy was much larger than could be accounted for by any conceivable chemical reaction the excess energy was attributed to an accompanying nuclear reaction. The calorimetric finding has been verified many times [2,3] but the claim that a nuclear reaction is the responsible agent remains repugnant to the majority of the nuclear physics community. It would be useful, therefore, to apply a technique that is more direct, simpler, and more transparent than calorimetry to confirm that a nuclear reaction can accompany electrolysis.

This paper is a review of the author's work since 2000 with plastic detectors applied to electrolysis. Energetic nuclear particles and very energetic neutrons can be detected and recorded by a high-polymeric material designated as CR39 that has been used by nuclear physicists [4] for many years. Upon entering the plastic, a nuclear particle produces a trail of disrupted chemical bonds which are more easily chemically attacked than undamaged material. The pit that results from etching the material in hot, concentrated alkali solution is unambiguous evidence that a nuclear reaction has taken place because the energies required to break the chemical bonds are much larger than can be provided by any chemical reaction.

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2. Experimental Procedure

The CR39 plastic used in this research was purchased with both sides covered by adhering plastic films designed to prevent the formation of nuclear tracks by air-borne radon. To prepare the CR39 for use in the experiments the desired chip sizes are obtained by scoring the plastic on one side with a sharp blade, and then cracking the plastic. Very small holes are drilled through the plastic-protected chips to enable their suspension within the electrolysis cell and also subsequently within the etching solution. If necessary, identifying characters are scratched on the chips through the protective film, which is removed just before mounting in the electrolysis cell.

The experiments consist in determining the number density of tracks that are produced in the electrolysis-exposed chips and comparing with the number densities produced in the chips used as controls. For this purpose, the experimental (active) and the control chips are etched together in stirred 6.5 N KOH or NaOH solution at 60–80°C for 6–24 h depending on the etchant temperature. After rinsing off the alkaline solution and drying, the etched chips are protected against radon either by covering with adhering Scotch tape or by tight wrapping in aluminum foil until examination under the microscope. The chips are examined immediately after cleaning, done by scrubbing with a small, soft brush in a detergent solution, rinsing under tap water, and drying in warm air. This procedure was found superior to ultrasonic cleaning.

To aid in the identification of nuclear-caused pits, experience was gained by examining etch pits caused by exposure to radiation from pitchblende and to ^{241}Am. Defects produced during the manufacture of the CR39 plastic can make it difficult to discriminate between nuclear pits and artifacts. The polymerization process can leave poorly polymerized regions. Such regions will etch more rapidly than does well polymerized material and pits will result. However, these pits are usually small, circular, and shallow, and can be distinguished from nuclear pits by the much darker appearance of the latter when examined under a microscope employing illumination from above. Features of dubious identity are not counted as nuclear pits.

Scratches and other mechanical insults to the detector chips can also cause etch pits. This problem was examined by a series of experiments in which various mechanical forces were applied to the chips, followed by etching and microscopic examination. Light scratching produces linear arrays of pits, usually of uniform diameter. For this reason, pits in linear arrays are disregarded. Etch pits are not caused by high pressure against the chip surface by the points of metal tweezers or by the handle of an Exacta knife. However, pressure with the point of an Exacta blade produces a very dark etch pit. Rubbing with a plastic rod has no effect, but rubbing with a metal spatula leaves a faint trace after etching, and rubbing with the points of tweezers produces an etchable trail. Grasping a detector chip with metal tweezers exerting a considerable force does not produce any effects.

The electrolysis apparatus consisted of a cylindrical glass tube of about 15 mm internal diameter whose lower end terminates in a flanged joint between whose O-rings can be clamped a cathode plate or a CR39 detector chip. For all of the experiments here reported, the electrolyte is a solution of lithium sulfate in either D_2O or distilled H_2O, the anode is a spiral of platinum wire, and the cathode is made of palladium, nickel, or platinum. The detector chips are examined at 100× and 500× magnification, and the etch pits are photographed in some of the experiments and are counted for all of the experiments. The details of cell design and operation differ between the types of experiments; these are presented in the sections below.

3. Electrolyses with Immersed Detectors

The electrolysis cell employs a platinum wire the upper end of which is spot-welded to a tungsten wire, and the lower end is fashioned into a spiral to serve as the anode. The palladium cathodes are 25 × 25 mm sheets of 1mm thickness obtained from Dr. E. Storms, Los Alamos Laboratory (retired). They are sanded and washed before being clamped between the Viton O-rings of the glass flanged joint. The nickel cathodes are of 99.9% purity and are of about 0.2 mm thickness. The electrolytes are a solution of 0.025 g of Li_2SO_4 per ml of D_2O or distilled H_2O. The electrolyses are

Table 1. Results of the Mann–Whitney analysis applied to the data for the immersed detectors

System	*P*-value
Pd/D_2O	2.5×10^{-5}
Pd/H_2O	1.2×10^{-6}
Ni/D_2O	5.5×10^{-4}

Note: *P* is the probability that the data for the active and for the control chips belong to the same population.

carried out with current densities between 0.1 and 0.37 A/cm^2 for two or three days with the evolved gases evolved into the environment. Chips of detector plastic are suspended within the electrolyte by platinum wires both above and below the anode spiral. To serve as controls one or two detector chips are immersed in electrolyte solution contained in bottles open to the air during the duration of the electrolysis, and are etched along with the active chips. These controls account for the exposure of all the detector chips to environmental radon during handling, etching, washing, and microscopic examination. Because bubbling within the electrolyte might increase the number of nuclear tracks caused by radon, additional non-electrolysis tests were performed in which D_2 gas bubbles, produced by forcing the gas through a fine fritted glass tube inserted in the solution, impinged upon detector chips suspended in the solution. The impingement of bubbles was found not to increase the number density of nuclear tracks over the number density found in chips kept in quiescent solution. Prior to use in the experiments, the chips are etched and various areas of the chips are photographed at 100× magnification. After their use the chips are again etched and the same areas are again photographed and the etch pits are counted. The difference between the numbers of etch pits before and after use are divided by the areas photographed to yield the number densities of nuclear pits produced by electrolysis or, in the case of the controls, by immersion in the bottled electrolyte solution.

Experiments of this type were carried out with Pd cathodes and D_2O/Li_2SO_4 as the electrolyte [5], with Pd cathodes and H_2O/Li_2SO_4 as the electrolyte [6,7], and with Ni cathodes and D_2O/Li_2SO_4 as the electrolyte [6,7]. In all instances there is some overlap between the number density of nuclear tracks produced by electrolysis and those produced in the controls. Some electrolyses produced as few tracks as did some of the controls. Hence, a careful statistical analysis must be applied to the data. A conservative analysis is that of Mann and Whitney [8]. It has the considerable merit of not being dependent on the data having a Gaussian distribution. The results of this analysis are presented in Table 1, in which P is the probability that the data for the active chips and those for the controls constitute a single population. The analysis shows that the data for the active chips are a distinct and significantly different population from that of the control data. One can conclude with a very high confidence level that the excess track densities in the active chips are caused by a physical agent associated with electrolysis. The agent can not be nuclear particles originating at the electrode because the distance through the solution covering the electrode is much larger than the mean free path of charged particles through the solution. It must be concluded that the nuclear particles are generated close to the surfaces of the detector chips immersed in the solution.

4. Detector Chips above the Electrolyte

The electrolysis cell was modified to provide for detector chips suspended in the vapor above the level of the electrolyte. To mitigate the accumulation of liquid water on the chips, the vapor space of the cell is surrounded by a heating coil to maintain a temperature of about 60°C, and a closely fitting nickel disc is placed between the surface of the electrolyte and the chips hanging on hooks. For controls, detector chips are suspended above electrolyte solution in closed bottles, without electrolysis. In this instance investigation of the effect of bubbling within the solution upon the control chips was deemed not necessary because the nickel disc interposed between the solution surface and the suspended chips in

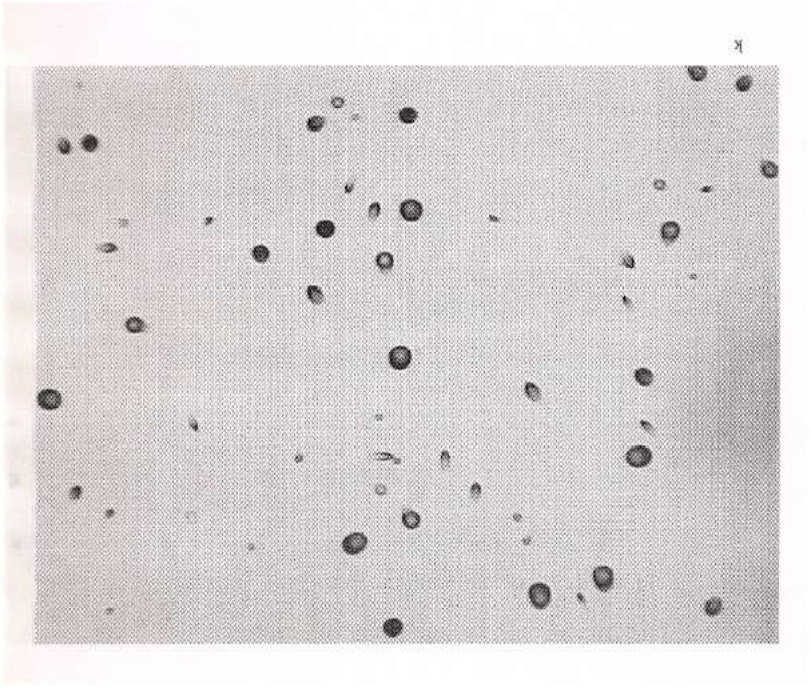

Figure 1. Nuclear pits produced on a CR39 chip suspended during electrolysis in the vapor above the solution of Li_2SO_4 in D_2O.

the electrolysis cell effectively prevented any effects from bubbling during electrolysis.

With either Pd or Ni as the cathode material and Li_2SO_4 in distilled H_2O as the electrolyte, most electrolyses produced number densities of nuclear pits exceeding the densities on the control chips [9]. The result of the Mann–Whitney test applied to the active and the control chips is $P = 3 \times 10^{-10}$, so that there is only a vanishingly small probability that the two sets of data constitute a single population. One may therefore conclude that electrolysis can generate nuclear particles by some kind of nuclear reaction that takes place in the vapor phase.

In some experiments very large numbers of nuclear pits were recorded. An example of what can be called showers is shown in Fig. 1. In one particularly interesting experiment, detector chips that during electrolysis had been suspended fairly close together in the vapor developed enormous numbers of nuclear tracks on the chip surfaces that had faced each other, and smaller numbers on their opposite sides. The etch pits on one side of these chips were laboriously counted to a total of about 33,000. From the shapes of the pits, and the angles and senses of the impingements of the responsible nuclear particles, the location of the point of origin of the nuclear particles was deduced to have been at about 2 mm from the chip surface in the vapor between the two chips.

5. Detectors Proximate to the Cathodes

It has been shown that detector chips immersed in, or suspended above, the electrolyte during electrolysis frequently develop many more nuclear tracks than do the control chips. However, not every experiment yields such pit densities. Hence it would be highly desirable to devise an experimental technique that yields a nuclear signature every time that the experiment is performed. Placing the detector chip very close to the cathode is the strategy adopted to try to achieve

Table 2. Summary of consecutive electrolysis experiments.

Exp. No.	Cell	Current (mA)	Duration (h)	Tracks/cm^2	
				Front	Rear
1[a]	S	0.2–25	168	284	150
2[a]	S	0.1–45	120	156	160
3[b]	S	12–100	102	—	—
4	S	70–143	67	352	16
5	S	30–75	96	393	498
6	S	50	97	76	74
7	S	5–50	96.5	71	96
8	S	10–100	94	80	70
9	S	300	65	D	D[c]
10	B	10, 25	98	98	40[c]
11	S	12, 28	97	229	48[c]
12	S	20	95	38[c]	167
13	B	27	95	193[c]	298
14	S	20	94	11	81
15	B	19	94	195	49
16	S	40	94	36[c]	9[c], 103[h]
17	B	39	94	127	9[c,h], 32[h]
18	S	60	93	28	102
19	B	60	93	47	35
20	S	80	117	72	41
21	B	80	117	60	132
22	S	100	93	426	207
23	B	101	93	62	51
24	S	50	96	102	38
25	B	50	96	26	344

[a]The electrolyte in these two experiments was LiCl plus $PdCl_2$ in D_2O with Ag as cathode material. All the other experiments used Li_2SO_4 in H_2O with Ni as cathode.
[b]The detector chip of Exp.3 was lost before a careful count of the clearly large number of tracks could be made.
[c]The chip area counted had been covered during electrolysis by the manufacturer-supplied blue plastic film. Track counting was done over the chip area subtended by the O-ring, 3.5 cm^2, except for items marked h, for which counting was over half of the 3.5 cm^2 area.
[d]Signifies that the number of tracks was so large that counting was impractical.

replicability. However, direct contact of the cathode with the detector runs the risk of chemical attack of the plastic by the hydroxyl ions produced by the reduction of water at the cathode. To preclude chemical attack, Mylar foil of 6 μm can be interposed between the electrolyte and the detector chip. Preliminary experiments were done to make sure that radiation from pitchblende can traverse the Mylar film. In the configuration adopted, a CR39 plate overlain by Mylar film is clamped between the O-rings of the glass ball joint and the lower end of the cathode rests upon the Mylar. The electrolyses usually employed a platinum anode and an electrolyte of lithium sulfate in distilled ordinary water. The electrolyses are followed by etching the active and the control chips and then counting the nuclear pits. For the actives the counting is restricted to the area circumscribed by the O-rings to avoid chip areas that might have been affected by radon during the electrolyses. Two different cells of identical design were employed for the 25 consecutive electrolyses, each of which lasted from two to four days.

The results of these experiments are presented in Table 2. The counts of nuclear tracks per unit area, either on the Mylar-covered surface (the front surface) or on the opposite surface, or on both surfaces, are always considerably

Table 3. Results of control experiments (Tracks/cm^2)

CR39 configuration	First CR39 sheet			Second CR39 sheet		
	N	Mean value	σ	N	Mean value	σ
Wrapped in Mylar	16	26.4	12.1	13	13.6	6.8
With new O-rings				10	16.2	6.0
In stock solution				14	5.9	2.7
In new cell				6	16.5	5.2

Notes: N is the number of tests.
σ is the standard deviation

greater than the counts on the control chips. The control experiments were of four kinds, each lasting two to four days. Some chips were wrapped in Mylar film, some immersed in electrolyte solution in which the working electrolyte had been stored, other chips were pressed against as-received O-rings and wrapped in aluminum foil, and other chips were mounted in a newly constructed cell fitted with unused electrodes, electrolyte, Mylar film and stoppers but without electrolysis. The results of the four kinds of controls using chips cut from two different CR39 sheets are presented in Table 3. A comparison of the active chips with the controls leads to the conclusion that a nuclear reaction of an unknown kind is consistently generated in the course of electrolysis. The many instances of nuclear tracks on the rear surfaces of the detector plates is particularly significant because it is strong evidence that ordinary radionuclides contaminating the electrolyte can not have been responsible for the observed tracks. This is because charged particles of 15 MeV or less can not traverse the plastic of 0.83 mm thickness.

It is worth remarking that the nuclear pits often appear in two distribution modes. One is a random scatter over the surface of the detector chip. The other mode is that of a cluster of nuclear pits surrounded by areas relatively devoid of pits. The clusters themselves are of two kinds: one in which the pits are randomly distributed as to position and shape, and the other in which the elliptical or conical axes of the pits are radially distributed. Figure 2 shows an example of the latter kind of cluster that indicates that the nuclear particles that produced the cluster originated at a common point. Clusters of two to five tracks diverging from one point also sometimes appear. These track patterns should prove helpful in the elucidation of the phenomena here described.

6. Internal Tracks

The finding of nuclear tracks on the rear surfaces of detector chips held between the O-rings of the electrolysis cell led to investigating whether or not nuclear tracks can be initiated totally within the thickness of the plastic. This question was addressed by the simple technique of repeated etching [10]. The first etching after electrolysis reveals the nuclear tracks generated by nuclear particles that entered through the original surface of the plastic. If a second etching removes a thickness of the plastic greater than the mean free paths of the nuclear particles and many new tracks are thereby revealed, more than can be accounted by radiation from radon in the air, then one can conclude that the majority of the second set of tracks was initiated within the original thickness of the detector. The effect of radon can be evaluated by conducting suitable controls, namely, counting the pits after the first and then after the second etching of chips never exposed to electrolysis.

Table 4 collects the results of the re-etching of detector chips that had been covered with Mylar and that had been clamped between O-rings during electrolysis. The increments in number densities of tracks after the second etch are to be compared with the results from the controls: an average of 5.6 new tracks per cm^2 after the second etch, ranging between 0 and 21 per cm^2; this reflects only the effect of air-borne radon. Thus, the electrolysis chips contain damage trails that begin below the depth in the plastic reached by the first etch after electrolysis. This is about 30 μm. These

nuclear pits could not have been the result of contaminating radionuclides external to the chip. The pits that appear only after the second etching of the electrolysis-exposed chips exhibit distributions of size and shape different from those that appear after the first etch.

7. Detectors in the Anode Compartment

The conventional thinking in the field of low energy nuclear reactions is that the reaction is produced at the cathode where deuterium from the electrolysis of heavy water dissolves at very high concentration in the palladium cathode. Of importance in the elucidation of the mechanism of the nuclear reaction that can accompany electrolysis would be the establishment of whether or not the cathodic reaction is essential. This question can be examined by placing CR39 detectors in the anode compartment of an electrolysis cell constructed in the form of a U in which platinum spirals in each leg of the U serve as the electrodes. The electrolyte of Li_2SO_4 in distilled H_2O is contained in the lower portions of the U-tube, the liquid extending sufficiently high into each leg so that the gases produced at the electrodes can not mix with each other. Detector chips are suspended above the liquid level in the anode compartment where they are maintained at about 60°C by an external Nichrome wire heater. Prior to electrolysis the detector chips are etched and examined for pre-existing nuclear tracks. Electrolysis with 0.1–0.4 A/cm^2 is done for two or three days, after which the chips are again etched and examined. For controls, detector chips are suspended above bottled electrolyte solution for two to four days without electrolysis. An additional precaution was to ascertain that holding detector chips in oxygen at 60°C does not damage the detectors.

Tracks were produced on chips held in the vapor in the cathode compartment. But more to the point, tracks were

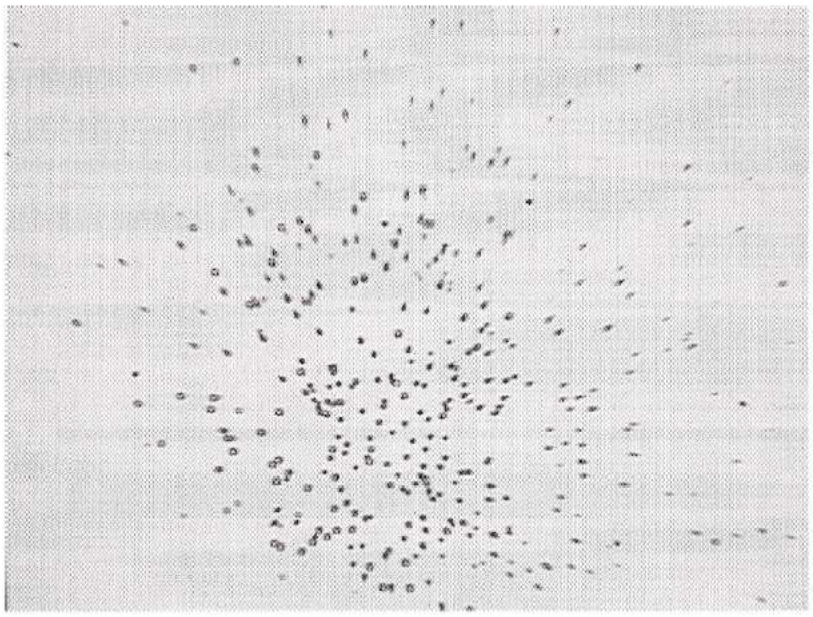

Figure 2. A cluster of nuclear pits formed on the rear surface of a CR39 chip. The angular shape of the pits and their distribution indicate that the nuclear particles that produced the nuclear tracks originated at a common point.

Table 4. Change of track count (tracks/cm^2) upon re-etching

After first etch	After re-etch	Increment	D (μm)
82	160	78	—
71	75	4	—
147	151	4	—
55	TL	TL	—
48	TL	TL	—
22	56	34	—
8	194	186	—
38	295	255	60
64	70	6	60
49	121	72	135
53	TL	TL	70
11	700	690	50
17	53	36	45
44	551	515	75
19	41	22	—
50	86	36	—
52	620	568	70

Notes: TL signifies that the number of tracks was so large that counting was impractical. D is the depth of recession of the chip surface from its original position caused by the etchings.

also produced on the CR39 chips suspended in the oxygen-water vapor mixture in the anode compartment. Fig. 3 shows an example of these nuclear pits. Table 5 summarizes the U-cell experiments that resulted in number densities of nuclear tracks much greater than those produced in the control chips. An equal number of U-cell experiments produced track densities comparable to the results with the control chips. These unsuccessful experiments provide assurance that ordinary physico-chemical phenomena associated with the moving oxygen gas are not responsible for the large track densities listed in Table 5. It is clear from these experiments that hydrogen-producing cathodic processes are not essential for causing the nuclear reaction that the CR39 plastic records.

8. Discussion

The relatively simple and transparent technique of using CR39 detectors has clearly shown that nuclear reactions of currently unknown mechanism can be produced during electrolysis. An experimental protocol has been developed that

Table 5. Detectors in the anode vapor space

Experiment	Front surface (tracks/cm^2)	Rear surface (tracks/cm^2)
1	253	349
2	306	250
3	272	301
4	258	307
5	612	385
6	238	200
7	326	228
8	300	245

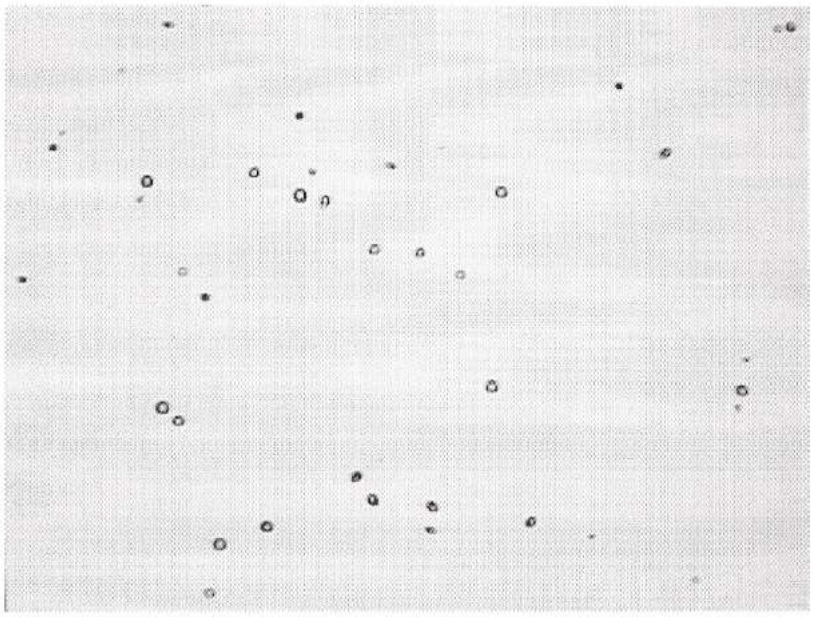

Figure 3. Pits formed on a chip suspended during electrolysis in the vapor in the anode compartment of the electrolysis cell where only oxygen is evolved.

is able reproducibly to generate the nuclear reaction. However, what specific feature of electrolysis is responsible for making the nuclear reaction possible is not at all clear. It has been shown that the use of heavy water with palladium as the cathode is not essential. Using light water and nickel cathodes is virtually equally effective, but what ions in the electrolyte are necessary is not known. Nuclear particles that leave damage trails in the detector plastic can be produced within the electrolyte, and in the vapor space that contains either hydrogen and water vapor or oxygen plus water vapor. More surprising is that nuclear particles can be generated within the thickness of the plastic. Any attempt to develop the mechanism of the nuclear reaction involved should take the phenomena here described into consideration. At present there is nothing in nuclear physics as currently understood that can account for these experimental facts. It is clear that the nuclear reaction that produces the tracks recorded by the CR39 detectors is not the cause of the anomalous excess energy that has been measured calorimetrically by many investigators beginning with Fleischmann et al. [1]. Nevertheless the mechanism of the track-producing nuclear reaction needs to be understood in order to progress towards developing a much needed non-polluting source of nuclear energy.

Acknowledgment

Much appreciation is expressed for valuable discussions with John C. Fisher, whose theoretical developments [11] inspired and supported some of the experimental work here described.

References

[1] M. Fleischmann, S. Ponns and M. Hawkins, *J. Electroanal. Chem.* **261** (1989) 301.
[2] R.A. Oriani, J.C. Nelson, Lee Sung-Kyun and J.H. Broadhurst, *Fusion Techol.* **80** (1990) 652.
[3] E. Storms, *Proc. 8th Intern. Conf. Cold Fusion*, 2000, p. 55.
[4] R.L. Fleischer, P.B. Price and R.M. Walker, *Tracks in Solids*, University of California Press, Berkeley, CA.
[5] R.A. Oriani and J.C. Fisher, *J. Jpn. Appl. Phys.* Part 1 **41**(10) (2002) 6180; Erratum, ibid. **42**(3) (2003) 149.
[6] R.A. Oriani, *Trans. Amer. Nucl. Soc.* **88** (2003) 640.
[7] R.A. Oriani and J.C. Fisher, *Proc. 10th Intern. Conf. Cold Fusion* , 2006, p. 577.
[8] F. Mosteller and R.E.K. Rourke, *Sturdy Statistics: NonParametric Order Statistics*, Addison–Wesley, Reading, MA, 1975 .
[9] R.A. Oriani and J.C. Fisher, *Proc. 10th Intern. Conf. Cold Fusion*, 2006, 567.
[10] R.A. Oriani and J.C. Fisher, *Proc. 11th Intern. Conf. Cold Fusion*, 2006, 295.
[11] J.C. Fisher, *Fusion Techol.* **34** (1992) 66.