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Reproducible Evidence for the Generation of a Nuclear Reaction During Electrolysis

R. A. Oriani
University of Minnesota, 151 Amundson Hall, 421 Washington Ave. SE
Minneapolis, MN 55455

ABSTRACT

Past work in this laboratory has shown that nuclear particles generated during electrolysis can be registered by CR39 plastic detectors held within the electrolyte solution, suspended in the vapor above the solution, or placed just below the metal cathode that serves as the bottom of the electrolyte compartment of the electrolysis cell. However, not every electrolysis experiment produced nuclear particles so that total reproducibility was not achieved. Therefore another experimental technique has been developed which has shown the generation of nuclear particles in each of twenty five consecutive electrolysis experiments using heavy or light water solutions of lithium salts. The damage trails caused by the nuclear particles are made visible by etching in hot concentrated caustic solution, and the electrolysis experiments are accompanied by suitable blank, or control, experiments. The damage trails begin either at the surface of the CR39 chip that faces toward the electrolyte, at the opposite surface, or totally within the 0.83 mm thickness of the plastic detectors. It is demonstrated that the nuclear damage trails could not have been caused by ordinary radionuclides contaminating anything involved in the experimental procedure. The described phenomena pose a formidable challenge to nuclear theory.

1. INTRODUCTION

Energetic charged particles and very energetic neutrons can be detected and recorded by a high polymeric material called CR39, and its use is well known in the nuclear community [1]. Upon entering the plastic, a nuclear particle leaves a trail of disrupted chemical bonds which are more easily chemically attacked than those in undamaged material. The pit that results from etching by concentrated alkali solution is unambiguous evidence that a nuclear reaction has occurred because the energies required for such damage are much larger than can be provided by chemical reactions.

The integrating character and the relative simplicity of this technique have led the writer to use it within electrolysis cells. Control, or blank, experiments are necessary because radon in the air can produce etchable damage trails in CR39 detectors. Past work with this method in this laboratory has included immersion of CR39 chips in electrolyte solution, suspension of the chips in the vapor above the solution, and placement of chips in air just below the metal plate that serves both as cathode and as the bottom of the electrolyte compartment, with each of these configurations maintained during electrolysis. The count of nuclear tracks (pits) on the detector chips used in the electrolyses and the pit counts on chips used as controls can be used to calculate

the probability, P, that the two sets of pits belong to the same population, i.e., that the electrolysis has no effect on the number of pits. The results of prior research [2,3] are as follows. For detector chips immersed in H_2O/Li_2SO_4 electrolyte and with Pd as cathode $P = 1.2 \times 10^{-6}$, and with Ni as cathode $P=5.8 \times 10^{-4}$. With D_2O/Li_2SO_4 and Pd as cathode $P = 2.5 \times 10^{-5}$. With detector chips suspended in the vapor above H_2O/Li_2SO_4 electrolyte and Ni or Pd as cathode material $P=3.0 \times 10^{-10}$. Although these results reasonably demonstrate that electrolysis can indeed generate nuclear particles, it can not be claimed that every electrolysis experiment will produce the nuclear reaction since the data sets for the electrolysis chips and the control chips partially overlap. Consistent reproducibility was not achieved despite having kept constant all the controllable parameters. Therefore a different experimental approach has been adopted.

2. EXPERIMENTAL DETAILS

Investigators [4] at the SPAWAR Systems Center, San Diego California, initiated the strategy of having the cathode very proximate to the CR39 chip. The metal wire serving as cathode was wound tightly around the detector chip. Unfortunately, this configuration results in copious chemical attack on the detector plastic by ions generated by the electrochemical reactions at the cathode. The huge number of chemical pits produced makes it very difficult to verify the generation of nuclear pits if any. To maintain the desirable nearness of cathode to detector while avoiding the chemical attack, the obvious modification is the interposition of a thin Mylar film between the electrolyte and the detector chip. Preliminary experiments proved that 6 □m Mylar film permits the passage of nuclear particles emitted by pitchblende, initial energies of 4.1 to 5.8 MeV.

The cell design is shown in Fig. 1. Squares of CR39, roughly 3 cm × 3 cm, cut from a sheet obtained from the Landauer Corp. are overlain by the Mylar films obtained from the Cemplex Industries, Inc., and the combination is pressed between Viton o-rings that fit into grooves in the glass o-ring joint purchased from the Fisher Scientific Corp. The joint is held together by a pinch clamp. The anode is a platinum wire the lower end of which forms a loop parallel to the plane of the detector chip. The cathode is usually a nickel wire whose lower end is bent into the shape of a W, the plane of which is parallel to the plane of the detector square. The vertical portion of the nickel wire is sheathed in heat-shrinkable plastic tubing and spot-welded to a titanium rod inserted in a glass tube. The electrode assembly is held together by a rubber stopper through which a hole permits the escape of the gases produced by the electrolysis. This describes the electrolysis cell designated by S. Another cell, constructed to enable more experiments to be done per week and designated by B, differs from the S-cell only in that the platinum anode wire ends in a crude spiral the plane of which is perpendicular to the plane of the detector chip.

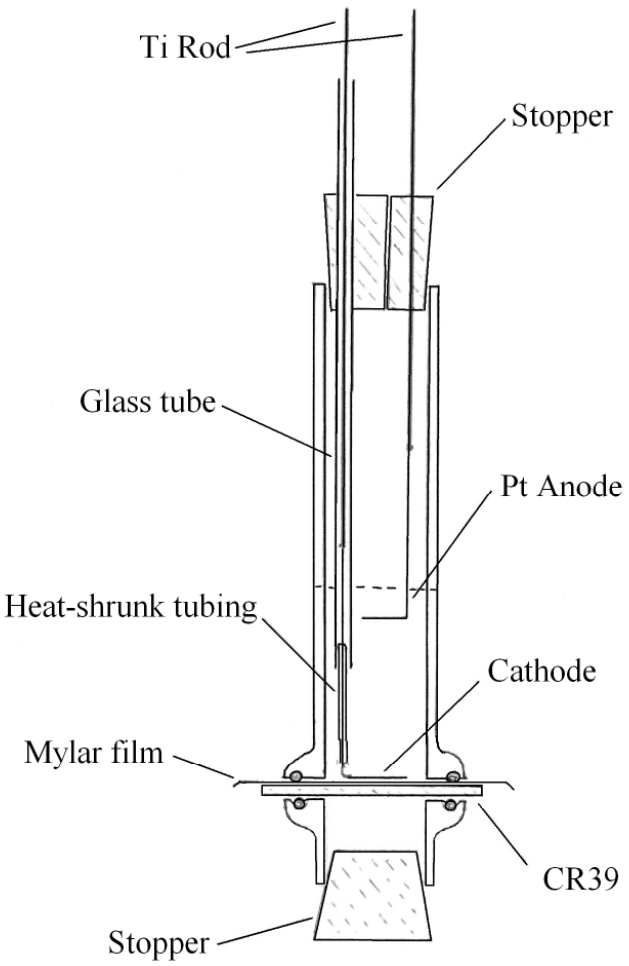


Figure 1. The electrolysis cell. The cathode assembly can be gently slid up and down to contact the Mylar film lying upon the detector chip.

The current to the S-cell was supplied and controlled by a potentiostat, and that to the B-cell by a constant-current power supply. To begin an experiment a square piece of CR39 is cut from the sheet from the manufacturer, a small hole is drilled at a corner of the detector chip to accommodate a nickel wire for the subsequent suspension of the chip, and an identification symbol is inscribed at another corner. The surface upon which the identifier is scratched is referred to as the front surface and is the surface upon which the Mylar foil is laid after the manufacturer-supplied blue protective film is removed. After the detector with the overlying Mylar film is clamped in the o-ring joint about 10 ml of electrolyte solution are poured into the cell and the electrode assembly is put in place. The cathode is then carefully lowered by sliding the supporting titanium rod within the supporting glass tube until the W-shaped foot of the cathode rests upon the Mylar film. Electrolysis is then started and the current is either kept constant for the entire duration of the electrolysis, or is increased once a day. The electrolyte employed was usually of distilled H_2O with 0.022 g Li_2SO_4 per ml. To provide a comparison with the results of the SPAWAR team the first two experiments were done with silver cathodes

in electrolyte of D_2O with LiCl plus $PdCl_2$. Water was not added during the electrolysis to compensate for the loss due to dissociation. Four days of electrolysis was the usual duration, after which the cell was disassembled and the detector chip removed and washed. There was never any trace of pitting or abrasion on the detector chip before etching. Stirred sodium hydroxide solution of 6.5 M initial concentration was used as the etchant for various etching times at temperatures of 70°C or higher. This was followed by examination at 100X and 500X, applying experience gained from examining pits produced by exposure of CR39 to ^{241}Am and to pitchblende to distinguish nuclear pits from artifacts caused by manufacturing defects in the detector plastic. The nuclear pits were counted on the surface of the detector chip overlain by the Mylar foil (the front surface) as well as on the opposite surface (the rear surface). The pits were counted only within the area bounded by the outer perimeter of the o-ring, an area amounting to 4.5 cm^2. This procedure avoided the counting of tracks that may have been caused by air-borne radon during the duration of electrolysis.

Because it was found that nuclear pits are produced on both sides of the detector chips, the possibility was investigated that nuclear tracks can be produced wholly within the thickness of the detector, i.e., that damage trails begin within the interior of the plastic. After completion of the electrolyses the chips were etched and examined at 100X magnification and the etch pits were counted. A second etching and microscopic examination were then carried out in which the etch pits were again counted. In a few experiments some ten markings were scratched on the front surfaces of the detector chips after the first etch, and the marked regions were photographed under the microscope. After the second etching exactly the same marked regions were again photographed. Efforts were made to protect the chips from exposure to air as much as possible during the entire course of the experiment. Between etchings the chips were kept tightly wrapped in aluminum foil.

3. CONTROLS AND POSSIBLE ARTIFACTS

Control experiments of four kinds were carried out to account for the nuclear tracks already present in the detectors as received from the supplier, as well as those produced during the entire experimental process by radon in the air, in the electrolyte, in the etching solution, and in the wash water. The control detector chips were handled in exactly the same way as were the experimental chips except that instead of being used in electrolysis they were either wrapped in Mylar film, immersed in stock electrolyte solution, pressed against as-received o-rings, or mounted in a newly constructed cell fitted with unused electrodes, electrolyte, Mylar film and stoppers, but in the absence of electrolysis. In each instance the same length of time as the electrolysis experiment was used. The results for the controls are presented in Table I. The number density of tracks in the as-received detector sheet varies from one shipment to another, and is not a constant for any one sheet. Therefore, it is appropriate to compare an experimental value of track number density with the mean value for the controls with due regard for the range of values for the controls. Electrolysis causes bubble generation which in turn causes convection currents that may bring daughter products from radon into close proximity to the CR39 detector, causing a larger number of etch pits than found in CR39 chips immersed in quiescent electrolyte solution. This possibility was examined by injecting bubbles of H_2 or D_2 gas through a fritted glass sparger into H_2O/Li_2SO_4 solutions containing CR39 chips for periods of times of three or four days. After etching it was found that the chips did not display any statistically significant difference from the results obtained with quiescent electrolyte solution shown in Table I.

TABLE I. Results of Control Experiments

METHOD	FIRST SHEET				SECOND SHEET			
	N	RANGE	MEAN	σ	N	RANGE	MEAN	σ
CR39 in Mylar	16	7.6 - 47	26.4	12.1	13	5.3 - 31	13.6	6.8
New O - ring					10	4.9 - 20.7	16.2	6.0
Stock solution					14	3.3 - 11.7	5.9	2.7
In new cell					6	8.8 - 24.7	16.5	5.2

NOTES:
N = number of tests
σ = standard deviation

Controls for the experiments that looked for internally generated nuclear tracks were of two kinds. One of these consisted in mounting detector chips and Mylar films in a cell, a twin of the electrolysis cell that had never been used for electrolysis. The "virgin" cell was filled with $H2O/Li_2SO_4$ solution from the same stock used for the electrolysis experiments, and was fitted with new Pt and Ni electrodes and new o-rings. Detector chips remained in the "virgin" cell without ongoing electrolysis for the same length of time as the electrolyses lasted in the experimental cell, after which the chips were etched, the etch pits examined and counted, and the chips re-etched and the resulting etch pits again counted. The second control experiment consisted in etching CR39 chips, counting the etch pits, and after a few days re-etching and again counting the pits. The two kinds of controls produced about the same increase in number density of pits between the two etchings, about 10 per cm^2.

Consideration has been given to the possibility that the features seen after etching might have been caused by processes other than a nuclear reaction. For example, radioactive particles floating in the laboratory air may have adventitiously settled upon a detector chip and produced nuclear tracks. This possibility was examined by placing fine particles of pitchblende upon detector chips. Examination after etching showed that such particles produce "rosettes" of tracks. These track configurations, reputedly also produced by cosmic rays, have very occasionally appeared on detector chips during our research. They were not included in the counting of nuclear tracks. One may suspect that electrostatic charges produced by peeling off the manufacturer-supplied blue, protective plastic film from the CR39 might cause pits after etching. This possibility was explored by adhering Scotch tape to a detector chip then peeling it off. After etching nothing was visible that could be attributed to static charges. However, it is recognized that if after peeling off the protective film the bare chip is allowed to remain exposed to dry air for many hours, the electrostatic charges on the chip will attract daughter products of air-borne radon so that nuclear tracks would be generated. This was prevented in our work by always mounting the detector chip in the electrolysis cell immediately after peeling off the protective film.

Manufacturing defects can make it difficult to discriminate between nuclear pits and artifacts. The polymerization process can leave poorly polymerized regions. As etching proceeds, the receding surface of the detector chip intersects with such regions which etch more rapidly than does well polymerized material and pits result. These pits are usually small, circular, and shallow, and most of them appear in groups. They can be distinguished from nuclear pits by the

much darker appearance of the latter in the microscope when illuminated from above. Features whose identity remains ambiguous were not counted as nuclear pits. Scratches and other mechanical insults to the detector chips can also produce ambiguities. This problem was examined by a series of experiments in which various mechanical forces were applied to chips, followed by etching and examination. Light scratching produces linear arrays of pits, usually of uniform diameter. For this reason pits in linear arrays were never counted as nuclear pits. Etch pits are not produced by hard pushes against the chip surface with the point of metal tweezers or with the handle of an Exacta knife. On the other hand, a push with the point on the 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; and rubbing with the point of tweezers produces an etchable trail. Grasping a detector chip with metal tweezers using considerable force does not produce any effects. This is fortunate because the chips are manipulated with tweezers.

4. RESULTS AND DISCUSSION

The primary purpose of the first series of experiments was to establish whether or not the present technique repeatedly produces evidence that a nuclear reaction can accompany electrolysis. Table II displays the results of 25 consecutive experiments in which the counts of nuclear tracks, either on the front or on the rear surfaces or on both surfaces, are always considerably greater than those for the controls (Table I). Hence it can be concluded that the present technique has consistently produced evidence that a nuclear reaction of some sort has been generated in the course of electrolysis. It is significant that the generation of nuclear tracks is not limited to the use of one electrolyte composition, to one kind of cathode metal, or to one value of electrolysis current. The control experiments with CR39 chips immersed in quiescent electrolyte solution (Table I) and in stock solution stirred by hydrogen bubble injection have demonstrated that the results shown in Table II for track number density on the front surfaces of the chips can not have been produced by radioactive contaminants distributed throughout the volume of the electrolyte.

The experimental results listed in Table II indicate instances of nuclear tracks developing on both sides of the detector chips that had been mounted in the operating electrolysis cell. Indeed, often the track number density on the rear surface is larger than that on the surface that faced toward the electrolyte. This is significant because it is further evidence that ordinary radionuclides contaminating the electrolyte could not have been responsible for the observed tracks. Charged particles that caused the tracks on the rear surfaces could not have been generated on the electrolysis side because the 0.83 mm thickness is much too large to be traversed by nuclear particles with energies of 15 MeV or less.

TABLE II. Summary of Consecutive Electrolysis Experiments. The first 11 experiments were done with the detector chips cut from the first CR39 sheet (see Table I), all of the rest were done with a second sheet.

Experiment	Cell	Current (mA)	Duration (hours)	Tracks/cm^2	
				Front	Rear
1[a]	S	0.2-25	168	284	150
2[a]	S	0.1-45	120	156[c]	160
3[b]	S	12-100	102	—	—
4	S	70-143	67	352	16
5	S	30-95	96	393[c]	498[c]
6	S	50	97	76	74
7	S	5-50	96.5	71[c]	96
8	S	10-100	94	80	70[c]
9	S	300	65	d	d
10	B	10, 25	98	98	40[e]
11	S	12, 28	97	229[c]	48[e]
12	S	20	95	38[e]	167[e], 163[e]
13	B	27	95	193[e]	298
14	S	20	94	11	81[c]
15	B	19	94	195	49[c,f]
16	S	40	94	36[e]	9[c], 103
17	B	39	94	127	9[c], 32[f]
18	S	60	93	28	102[c]
19	B	60	93	47	35[f]
20	S	80	117	72[c]	41[c]
21	B	80	117	60[c]	132[c]
22	S	100	93	426	207
23	B	101	93	62	51[f]
24	S	50	96	102	38[f]
25	B	50	96	26	344[f]

a. The electrolyte in these two experiments was LiCl plus PdCl. in D_2O with Ag as cathode material. For all other experiments Li_2SO_4 in H_2O with Ni as cathode was employed.
b. In experiment 3, the detector chip was lost before a careful count of the clearly large number of tracks could be made.
c. Some of the tracks appear in clusters.
d. The number of tracks was so great that counting was impractical.
e. The blue protective film supplied by the manufacturer was kept on the detector chip; in exps. 12, 16, and 17, only one-half of the detector surface was kept covered by the film.
f. D_2O was maintained within the closed air space below the detector chip.

It is interesting that the nuclear pits are distributed bimodally, both as individual randomly distributed pits and as dense groups or clusters that are surrounded by areas relatively devoid of

pits. The number densities in the clusters are much greater than the mean number densities listed in Table II. The clusters are of two kinds: one within which the pits are randomly distributed as to position and shape (Fig. 2), and one in which the elliptical or conical shape are radially distributed (Fig. 3). Pits of this shape result when the path of an impinging nuclear particle is other than perpendicular to the surface of the detector. By focusing the microscope up and down one can determine the direction and sense of the path [2]. Doing this for all of the elliptical or conical pits in the cluster one can determine that the nuclear particles that produced those pits emanated from a common origin away from the surface of the detector. In fact, by a careful analysis [5], one can estimate the distance above the detector surface where the shower of nuclear particles originated. Clusters such as the one shown in Fig. 3 can not be generated by a sequential decay of ordinary radionuclides dissolved in the electrolyte. Such a source of charged particles could not remain stationary in the convection currents caused by the bubbling during electrolysis long enough to produce a radial distribution of elliptical etch pits whose axes intersect at one common point. Such clusters furnish additional evidence that a nuclear reaction of unknown nature can develop during electrolysis.

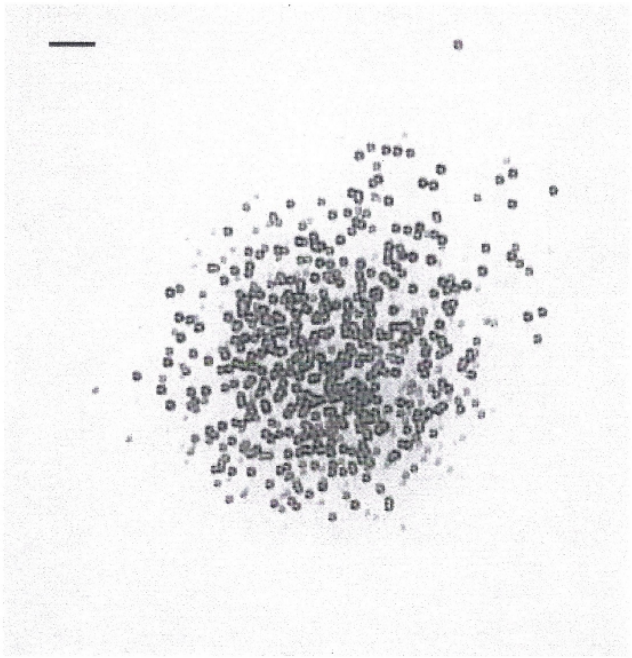


Figure 2. A typical group of etch pits here called a cluster. It is a high-density grouping of pits without any orientational arrangement among them, occupying an area of the detector chip otherwise fairly devoid of etch pits. (Scale line = 82 μm).

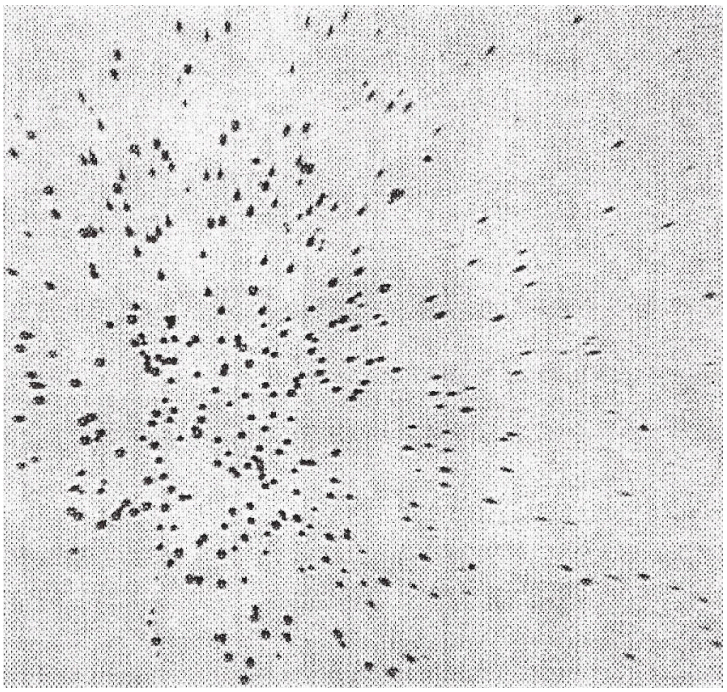


Figure 3. A radiating cluster typically containing pits of various shapes with the major axes of the non-circular pits radiating out from a common center. (Scale line=76 μm).

Further evidence that electrolysis can be accompanied by a nuclear process is provided by the re-etching experiments. The changes of etch pit counts on the detector chips that had been used in electrolysis experiments are shown in Table III. One sees that many of the re-etchings have produced large numbers of additional etch pits. The increases are to be compared with the pit count upon re-etching the control chips, about 10 pits/cm^2, reflecting only the effect of radon in the air, in the stock solution, and in the etching solution. The pits that appear after the second etch exhibit distributions of sizes and shapes very different from those of the pits that had appeared after the first etch. This is illustrated in Fig. 4. The large circular pit has resulted from the growth of a pit that was present after the first etch. The others appeared only after the second etch had caused the recession of the chip surface by 50 to 140 μm in from the original surface of the detector chip.

This latter set of pits could not have been the result of damage produced by nuclear particles that entered the plastic through the original surface of the detector chip because the mean free path of energetic protons and alpha particles in the plastic is considerably smaller than 50 □m. The lenticular shape of many of the etch pits show that the damage trails began many micrometers in from the original surface of the chip, so that the energetic charged particles that produced the damage trails must have been generated within the plastic material. Particularly interesting are the instances that show pitting of damage trails some 50 μm in from the original *rear* surfaces of the detector chips. These cases indicate that the energetic charged particles that produce the damage trails that lead to etch pits can originate at all depths within the 0.83 mm thickness of the plastic. This result is strong evidence against the hypothesis of contamination by radioisotopes in the electrolyte because the origins of the internal damage trails lie deeper within the plastic than the penetration depth of protons or alphas from radionuclides in the electrolyte.

TABLE III. Change Of Etch Pit Count Upon Re-Etching

Experiment	Chip Side	Pits per cm^2		Thickness upon re-etching (mm)
		After 1^{st} etch	After 2^{nd} etch	
1	F	82	160	-----
2	F	71	75	-----
3	F	147	151	-----
4	F	55	TL	-----
	R	48	TL	-----
5	F	22	56	-----
6	R	8	194	-----
7	R	8	67	-----
8	F	38	295	0.71
	R	64	70	0.71
9	F	49	121	0.56
10	F	53	TL	0.69
11	F	11	700*	0.73
12	F	17	53	0.74
13	F	44	551	0.64
14	F	19	41	-----
	R	50	86	-----

NOTES:
* Estimated
F, R: Front, Rear surface of the detector chip
TL: The number of pits is too large for convenient counting
The original thickness of the detector chips is 0.83 mm.
The mean number of pits/cm^2 of the controls after re-etching is 10 with a range of 0 to 21 in 18 tests.

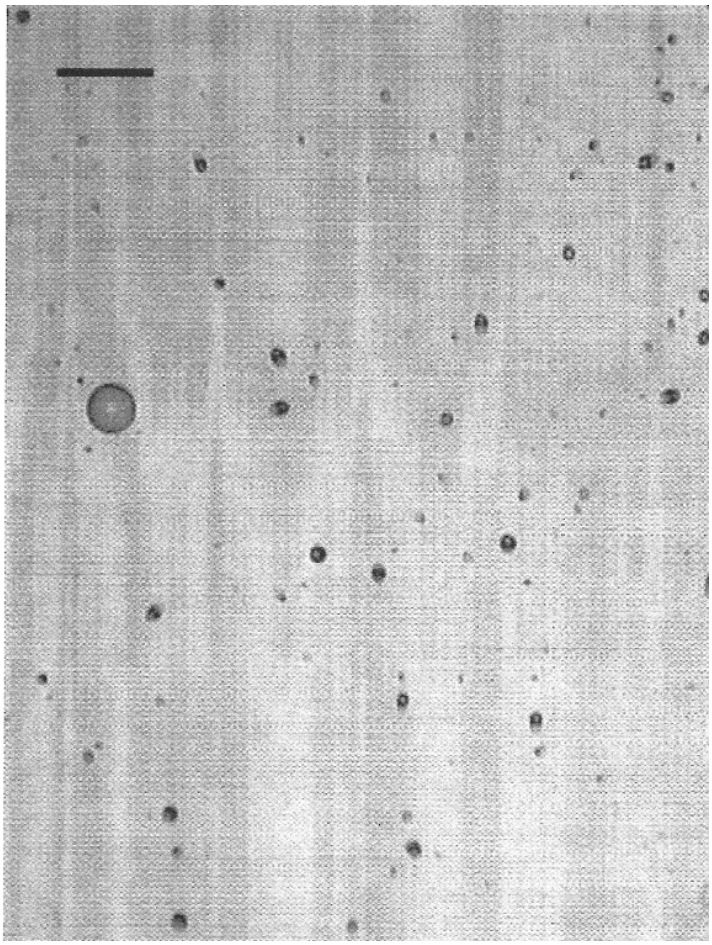
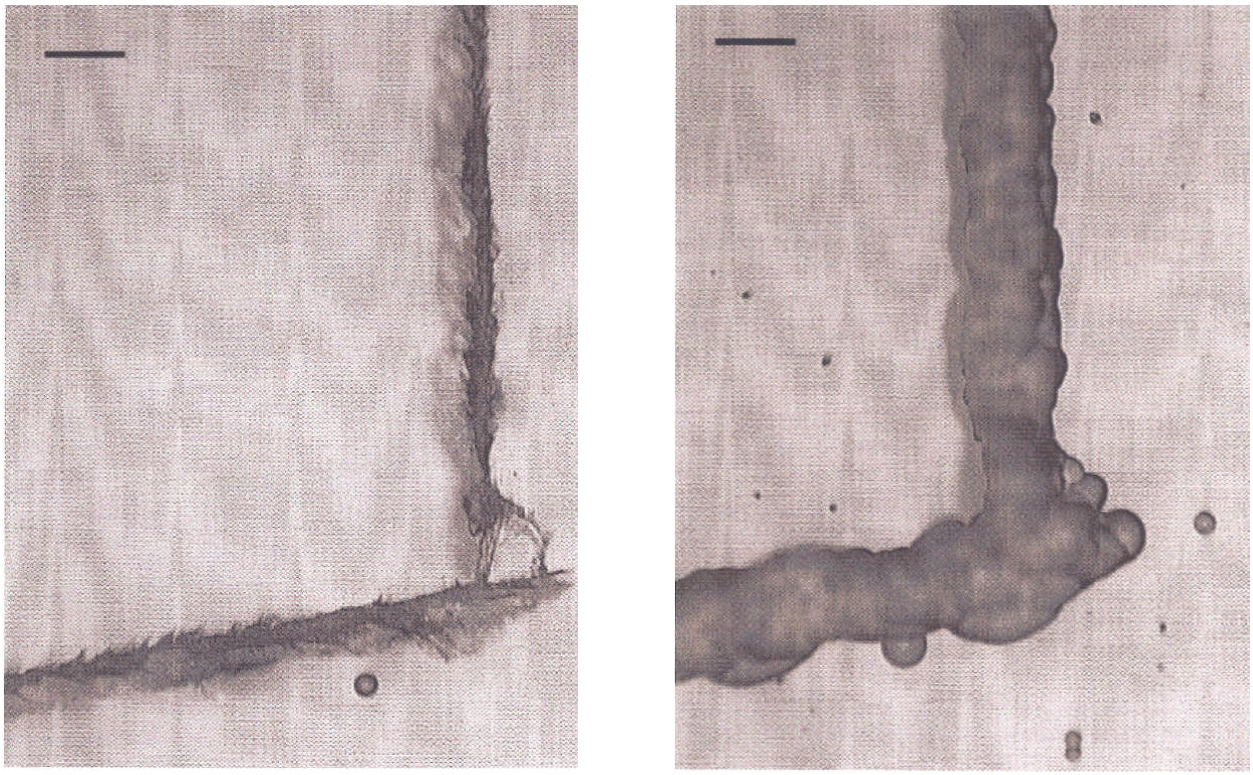


Figure 4. Photomicrograph (Scale line= 80 μm) of the front surface of a CR39 chip previously employed as a detector of nuclear particles generated during electrolysis. This chip showed 43.7 etch pits per cm^2 after the first etch and 551 after the subsequent etch. The second etch produced numerous smaller pits, in strong contrast with the single, large, circular pit made large by the re-etching of a pit already present after the first etch. The smaller pits appear classifiable into two families, probably associated with a distribution in depth of the starting points of the pits.

Several of the chips used in this study were marked after the first etch in order to be photographed at the marked locations both before and after the second etch. The marked chips showed large overall increases in pit count after the second etch, but the increases occurred in patches unevenly distributed over the area of the chip. Unfortunately, in only one case did one of these patches coincide with one of the marked and pre-photographed locations. Fig. 5a,b illustrates this instance. It is clear in Table III that not every second etch resulted in displaying internally generated damage trails. Either the phenomenon is of stochastic character, or all of the experimental parameters are not being controlled due to ignorance of their nature.



a b

Figure 5a. Photomicrograph (Scale line= 80 μm) of a region of a CR39 chip after the first etch. The L-shaped figure was scratched after the first etch in order to be able to locate the same region after re-etching. Only one pit appeared after the first etch. 5b. Photograph of the same region after the second etch. The scratch mark has thickened and almost covers the original single pit, and a new family of pits has appeared, produced by the second etch.

5. CONCLUSIONS

A technique has been developed that consistently produces evidence that a nuclear process can accompany electrolysis of solutions of lithium salts in either heavy or light water. The evidence is in the form of nuclear damage trails made visible by the etching of CR39 plastic chips. The damage trails can begin at either external surface of the chip placed within the cell during electrolysis as well as within the interior if the thickness of the chips. It is demonstrated that the nuclear damage trails could not have been caused by the decay of ordinary radionuclides contaminating anything in the experimental procedure. Rather, the nuclear damage is caused by a nuclear process of currently unknown nature.

The described experimental results can not be explained by nuclear physics as currently understood. Mechanistic interpretations should be the goal of future research. The aims of the present work have been only to develop an experimental technique that reproducibly shows that a nuclear process of a new kind can accompany electrolysis, to demonstrate some of the characteristics of the nuclear process, and to provide convincing evidence that would justify initiating intensive research in this new area of nuclear physics.

ACKNOWLEDGMENTS

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