

Research Article

Issues Related to Reproducibility in a CMNS Experiment

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Abstract

Unexplained emission of charged nuclear projectiles due to electrolysis has been reported by Richard Oriani. Experimental results were said to be highly reproducible. Working independently, we were not able to observe emission of charged nuclear particles (in a chemical process similar to Oriani's) and therefore are unable to provide supporting evidence that the effect is reproducible.

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1. Introduction

Radioactivity, such as emission of alpha particles, has long been thought to be independent of chemical processes, such as electrolysis. The same is true for nuclear reactions including fusion and fission. The reason for this is that the kinetic energies of molecules in a typical chemical process are too small to overcome the Coulomb repulsion between positively charged nuclear particles [1]. This expectation was challenged by Pons and Fleischman [2], Oriani [3, 4], and other scientists, as described by Storms [5]. We were attracted by Oriani's work because his findings were impressive

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(a nuclear process taking place during electrolysis) and his methodology was simple. Our goal was to either confirm or refute the claimed reproducibility of the effect.

The electrolyte in Oriani's cell, Li_2SO_4 , is usually dissolved in light water at the concentration of 0.022 g per cubic centimeter. The cathode electrode is usually nickel while the anode electrode is usually platinum. To detect charged nuclear particles, Oriani uses CR-39 chips. This method of detection is widely used in personal neutron dosimeters as well as in other fields of research [6] such as measuring the amount of radon in the air and studying thermonuclear reactions. In Oriani's 2003 experiment, described in [3], the detector was suspended in the electrolyte between the anode and the cathode. The mean track density on the experimental chips was 470 tr/cm^2 while the mean track density on control chips was 168 tr/cm^2 . The corresponding standard deviations were 384 and 99 tr/cm^2 , respectively.

Referring to such results, Oriani et al. concluded in 2003 that: "a nuclear reaction of as-yet unknown nature can accompany the electrolysis." Experimental results, according to Oriani's more recent 2008 paper [4], are now highly reproducible. The purpose of the present study was to verify this assertion.

2. Oriani's Cell and New Experimental Results

The cell used in Oriani's new study [4] is shown in Fig. 1. It was similar to the cell used in [3]. The essential difference was the addition of a thin Mylar film ($6 \mu\text{m}$) placed between the CR-39 detector and the nickel cathode. That film was chosen to protect the CR-39 detector from the electrolyte without interfering with detection of expected nuclear particles.

Twenty-one experiments were performed and 42 CR-39 surfaces were examined, as described in his 2008 paper [6]. The track density on one of these surfaces was reported as "too large to count." Track densities on the remaining 41 experimental surfaces ranged from 9 to 498 tr/cm^2 , as illustrated in Fig. 2. The histogram was drawn without making a distinction between surfaces facing the cathode and surfaces facing away from the cathode. Note that the distribution of densities is not bell-shaped. Track densities on control CR-39 chips were usually much lower (mean 21 tr/cm^2 and standard deviation 9.7 tr/cm^2) than on most experimental chips (mean 122 tr/cm^2 and standard deviation 124 tr/cm^2). Mean track densities on surfaces facing the cathode, according to data published in [4], were essentially the same as mean track densities on surfaces facing away from the cathode.

On the basis of these observations Oriani concluded that "the present technique has consistently produced evidence that a nuclear reaction of some sort has been generated in the course of electrolysis." Investigations described in this paper were undertaken to determine if Oriani's results could be reproduced in another laboratory.

3. Our Cells and Experimental Results

Our experiments, done in four separate laboratories, were performed using cells that were similar to Oriani's cell. Figure 3 displays a generic diagram of our cells. We used the same electrolyte (Li_2SO_4 in distilled H_2O , at the initial concentration of 0.022 g/cm^3) as Oriani; our electrodes were also made from Ni and Pt. Oriani's nickel cathode wire and our nickel cathode wires were from the same spool. All of us used the $6 \mu\text{m}$ Mylar which is capable of transmitting alpha particles with energies higher than 2 MeV. Distances between the cathodes and the anodes were approximately 3 cm, which is approximately the same as Oriani's. The essential cell parameters of Oriani's and our experiments were the same.

Differences between Oriani's experiment and our experiment are the following: Platinum wires for the anodes did not come from the same manufacturer. Oriani's cathode was spot-welded to a vertical titanium rod, placed inside a glass tube. Vertical parts of our electrodes, on the other hand, were placed into long heat shrink tubes (not shown in Fig. 3). Lower parts of our shrink tubes were thermally compressed, to prevent capillary creep of the electrolyte. Also note that Oriani's cell exposes the bottom surface (the side away from the electrolyte) of the CR-39 detector to air while our

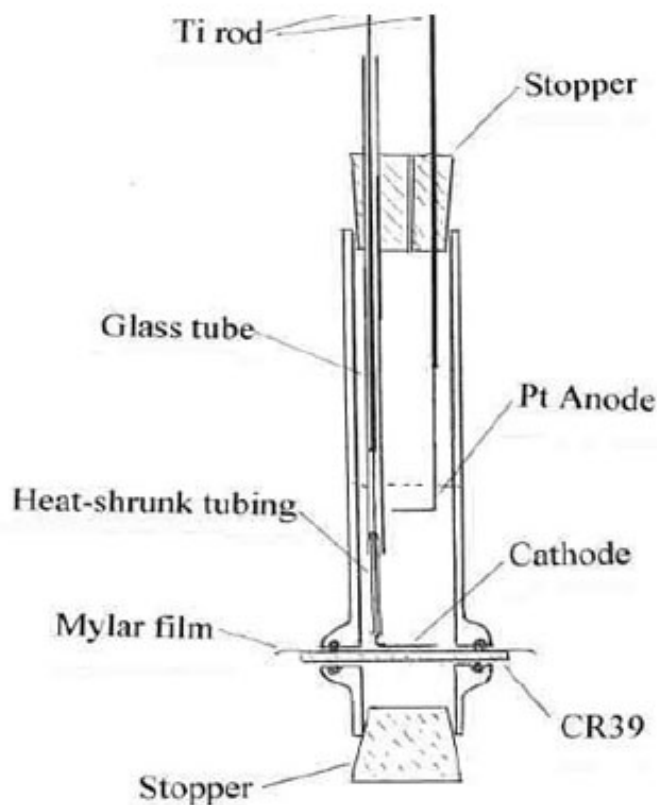


Figure 1. A diagram of an Oriani cell from his 2008 paper [4]. Small circles are cross sections of O-rings placed between the two sections of the cell. A clamp (*not shown*) is used to press the sections toward the CR-39 detector.

cells do not. In our cells, the bottom surface of the CR-39 is in contact with a 25" layer of polyethylene. That layer was clamped to the cell. The leakage of the electrolyte was prevented by squeezing the CR-39, Mylar and O-ring between the polyethylene and the cell.

Both we and Oriani used Fukuvi CR-39 material purchased from Landauer. The delivered sheets of that material are protected from scratches and alpha particles in air by a thin layer of polypropylene plastic. That layer usually peels off when the sheet is cut into small chips. Our unprotected chips were kept in salty distilled water (NaCl concentration of about 10 mg/cm^3). Furthermore, all control chips were kept in unused electrolyte during electrolysis. Salty water prevents potential accumulation of electric charges on CR-39 surfaces kept in air. This precaution was taken to reduce the possibility that electric charges on CR-39 surface in air might attract or repel radioactive ions. No chips were exposed to air for more than one hour. One author (P.L.) compared track densities on two CR-39 chips, one kept in

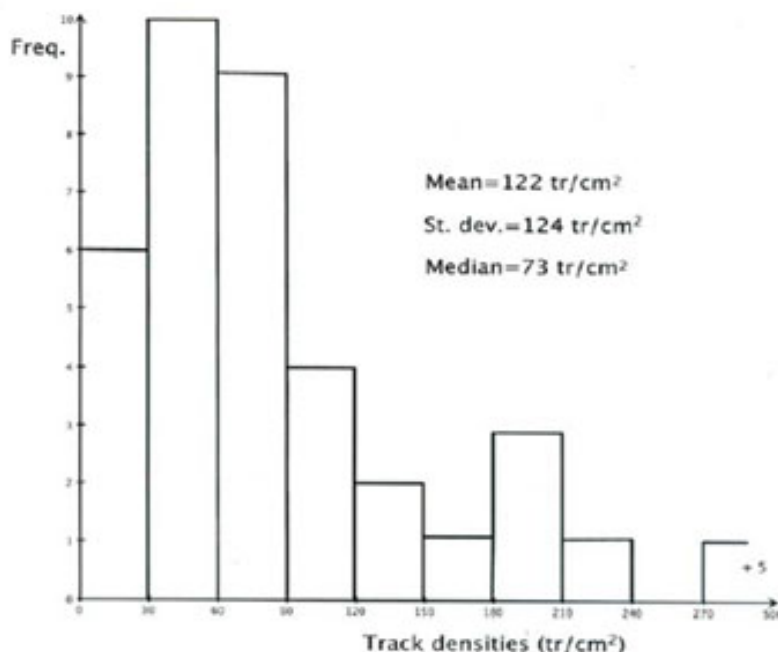


Figure 2. Oriani's distribution of track densities from his 2008 paper [4]. The +5 and 500, near the lower right corner, refer to five surfaces where track densities were between 300 and 500.

distilled salty water and in air, for 15 days. The track density turned out to be 30 tr/cm² on the chip kept in salty water and 505 tr/cm² on the chip kept in air. This can be explained by the big difference between ranges of alpha particles in air (centimeters) and in water (microns). Oriani did not use salty water; he wrapped the chips in aluminum foil and kept them in air. All of our “control” chips were kept in unused electrolyte during the time that the “experimental” chips were exposed to electrolysis.

Like in [4], we typically exposed the experimental chips to electrolysis for three days. During this time the electric current was changing slowly from 60 to about 90 mA, due to the loss of water. The corresponding voltage across the electrodes was typically 7–8 V and was kept constant during the experiment. The cell was dismantled after each experiment and the CR-39 chip was removed. It was then etched for 12 h in the stirred water solution of NaOH (concentration 6.5 M, temperature 72°C). The pits on CR-39 surfaces, identified as tracks of nuclear particles, were subsequently counted under the microscope. The uncertainty of up to plus or minus 5% (in reported track densities) was common due to difficulties in distinguishing tracks from certain surface defects. Each experimenter was given a CR-39 chip with one corner that was exposed to alpha particles from an ²⁴¹Am source. This helped the experimenter learn the difference between pits and defects. Pits due to particles from that source are shown in Fig. 4. They had the same size as pits found on control and experimental chips.

The mean and standard deviation on eight of P.L.'s experimental surfaces were 87 and 46 tr/cm². The mean and standard deviation on his five control surfaces were 59 and 38 tr/cm². This information, as illustrated in the Appendix, can be used to justify the statement that the reported difference between mean track densities on experimental and

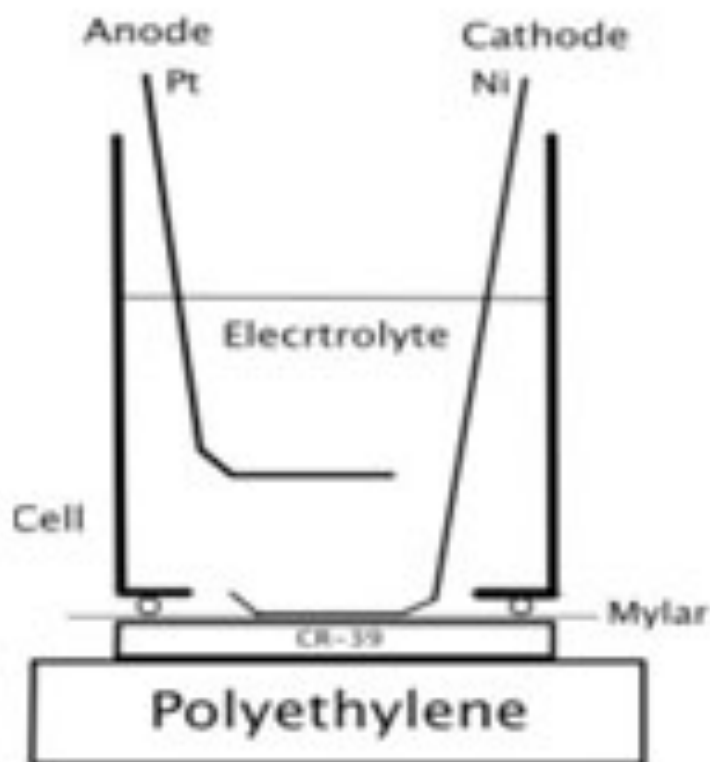


Figure 3. A generic diagram for our electrolytic cells. Small circles are cross sections of the compressed O-ring.

control surfaces, $87-59 = 28 \text{ tr/cm}^2$ was not statistically significant, considering small numbers of examined surfaces.

The mean and standard deviation on six of M.H.'s experimental surfaces were 185 and 73 tr/cm^2 . The mean and standard deviation on his eight control surfaces were 192 and 72 tr/cm^2 . This information was also used to justify the statement that the reported difference between mean track densities on experimental and control surfaces, $185-192 = -7 \text{ tr/cm}^2$ was not statistically significant.

The mean and standard deviation on eight of J.D.'s experimental surfaces were 98 and 17 tr/cm^2 . The mean and standard deviation on his six control surfaces were 125 and 29 tr/cm^2 , respectively. This information was used to justify the statement that the reported difference between mean track densities on experimental and control surfaces, $98-125 = -27 \text{ tr/cm}^2$ was not statistically significant.

The mean and standard deviation on nineteen of L.K.'s experimental surfaces (excluding three exceptional surfaces from experiments 13, 14 and 15 — see below) were 16 and 8 tr/cm^2 . The mean and standard deviation on his 16 control surfaces were 14 and 5 tr/cm^2 , respectively. This information was used to justify the statement that the reported

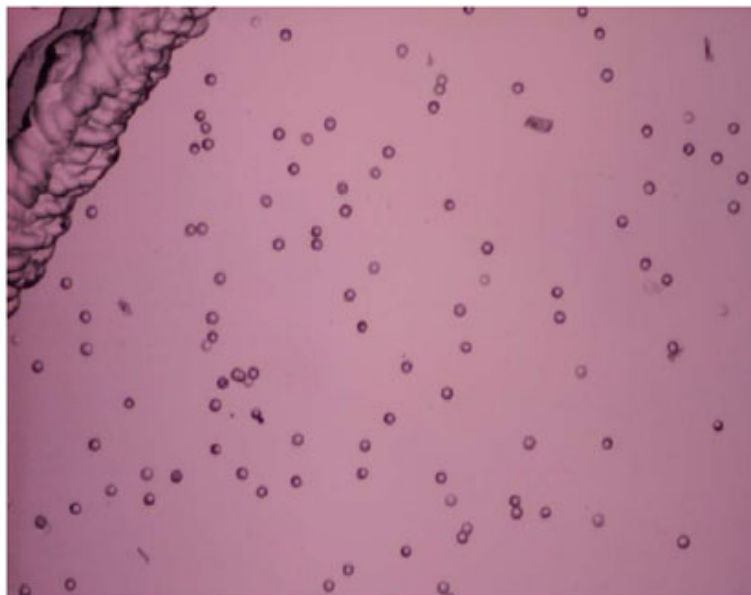


Figure 4. A CR-39 surface seen under the magnification of 40, after 12 h of etching. Small circles are pits due to alpha particles from an Americium source. The upper left corner shows a mechanically damaged area.

difference between mean track densities on experimental and control surfaces, $16-14 = 2 \text{ tr/cm}^2$ was not statistically significant. In other words, experimental results from our four sets of experiments are consistent with each other. But they are not consistent with the results reported by Oriani.

Let us address the issue of extremely high track densities on three surfaces from experiments 13, 14 and 15. The estimated track density on the up-facing surface of the CR-39 chip from Experiment No. 13 were around $15,000 \text{ tr/cm}^2$. This is two orders of magnitude higher than typical densities reported by Oriani [6]. The other side of the chip had nothing but the usual background. The area covered by copious tracks, nearly two square centimeters, matched the circle delimited by the O-ring. Distribution of tracks in that area was more or less uniform, except near the boundaries, where the track density decreased progressively.

Copious tracks from Experiment Nos. 14 and 15 were very different from Experiment No. 13. On these two surfaces, tracks were concentrated in small areas, near a hole drilled in each CR-39 chip. The holes were drilled to facilitate etching; each CR-39 chip was suspended in the etching solution by means of a thin copper wire threaded through the hole. These copper wires were from a telephone cable of unknown origin. The area covered by copious tracks from Experiment No. 14 was about 10 mm^2 ; the estimated track density was $200,000 \text{ tr/cm}^2$. The area covered by copious tracks from Experiment No. 15 was approximately 3 mm^2 ; the estimated track density was close to $12,000 \text{ tr/cm}^2$. Track densities outside of the small affected areas were about the same as on control chips.

It is remarkable that in each of the three cases of high track density, the mean density on the other side of the chip was essentially the same as on the control chips. That is very different from what was reported in [4]. The most likely cause of copious tracks was migrating radioactive contamination. Attempts made to identify it were unsuccessful. Alpha

radioactive substances such as uranium, thorium and radium are known to be present in our environment. One nanogram of radium, for example, emits 37 alpha particles per second. Testing of nuclear weapons in the 1960s contributed to contamination of our environment with long-lasting radioactive isotopes, tritium and uranium for example. Note that track densities on our three exceptional surfaces were much higher than those reported in [4].

Project participants worked independently of each other in four different states. Collective results, shown in Table 1, became available to participants only after their own results were submitted to L.K. He collected results after counting tracks on his own CR-39 surfaces. Note that Experiment No. 12 was performed without using the Mylar film. Low track densities resulting from this experiment seem to indicate that all experiments could have been performed without Mylar. Oriani used Mylar to protect the CR-39 from potential chemical effects. Most tracks on control chips were probably due to natural background, most likely due to radon, radium, etc. Concentrations of such substances are known to be location dependent. That fact was probably responsible for differences between mean track densities on the control chips of individual researchers.

4. Conclusion: Facts and Interpretations

The purpose of our four independent investigations was to find evidence for reproducible emission of nuclear particles described in Oriani's 2008 paper [4]. No such evidence was found. We examined 40 CR-39 surfaces and found only three cases of excessive tracks. No excessive tracks were found on the remaining 37 surfaces.

We do not know why we were not able to observe emission of charged nuclear particles (in a chemical process similar to Oriani's). There could have been a setup or procedural difference between the experiments. Further experimentation is needed to confirm the existence of nuclear particle emission in light water electrolysis, understand the causes of such

Table 1. Track densities on 34 surfaces of our experimental chips. No Mylar was used in Experiment No. 12.

Exp. No.	ID	Experimental chip (upper side) tr/cm ²	Experimental chip (lower side) tr/cm ²	Control chip averages tr/cm ²
1 P.L.	20	28		
2 P.L.	125	148		
3 P.L.	89	94		59 (from five surfaces)
4 P.L.	99	91		
5 M.H.	128	118		
6 M.H.	128	192		192 (from eight surfaces)
7 M.H.	278	267		
8 J.D.	121	113		125 (from six surfaces)
9 J.D.	87	85		
10 L.K.	21	23		
11 L.K.	24	24		
12 L.K.	18	8		
13 L.K.	> 10000	11		
14 L.K.	10	> 10000		
15 L.K.	> 10000	17		14 (from 16 surfaces)
16 L.K.	31	20		
17 L.K.	23	8		
18 L.K.	12	24		
19 L.K.	10	8		
cr 20 L.K.	11	3		

emission and determine the conditions required for reproducibility.

Appendix: An Example of Statistical Analysis

The mean and standard deviation on eight of P.L.'s experimental surfaces were 87 and 46 tr/cm². The mean and standard deviation on his five control surfaces were 59 and 38 tr/cm². Is the difference between mean track densities on experimental and control surfaces, $87 - 59 = 28$ tr/cm² statistically significant? To answer this question one must estimate uncertainties associated with actually measured track densities (87 and 59). The uncertainty about 87 is $46/2.8 = 16$, where 46 is the reported standard deviation while 2.8 is the square root of 8 (the sample size consisted of eight surfaces). Likewise, the uncertainty about 59 was $38/2.23 = 17$ tr/cm², where 2.23 is the square root of 5 (the sample size consisted of five surfaces).

The difference of 28 tr/cm² is too small to be significant, considering large uncertainties associated with the reported mean densities (59 and 38 tr/cm²).

The same approach was used to show that differences between mean values on experimental and control chips of -7, -27, and 2 tr/cm², as reported by M.H., J.D., and L.K. respectively, were also statistically insignificant.

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