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Research Article

Search for Low-energy X-ray and Particle Emissions from an Electrochemical Cell

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Abstract

Several theories to explain anomalous heat production predict the emission of low-energy X-rays and/or MeV alpha particles from PdD cathodes in electrochemical cells. Such radiation, however, is not detectable from outside of a standard electrochemical cell due to absorption in the electrolyte and cell walls. A custom cell was therefore assembled which permits X-rays of energy $\geq$ 1 keV to pass through a thin cathodic membrane and enter into an X-ray detector with minimal attenuation. This test cell geometry also potentially allows any emitted MeV alpha particles to be detected when they impact a Pd cathode and cause fluorescent emission of Pd-K_α (21.2, 23.8 keV) X-rays. The detection of X-ray emissions from a membrane electrolytic cell potentially permits the mechanism(s) for anomalous heat production to be investigated with great sensitivity. As an example, a typical X-ray detector allows 1 keV X-rays to be detected at emission rates of less than one per second and this level of sensitivity corresponds to a thermal resolution of < 0.2 fW. Time resolved X-ray spectral data ranging from 1 to 30 keV was collected for over a year using various types of membranes and different electrolytic solutions. None of these test cells, however, yielded any X-rays which were above ambient background levels.
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1. Introduction

Low energy emissions in electrochemical anomalous heat (AHE) experiments have never previously been thoroughly investigated by using time-resolved detectors with real time threshold analysis and continuous data collection. While

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Table 1. A list of various theories which predict low energy X-ray or particle emissions associated with AHE.

Author	Theoretical basis for emissions	Predicted emissions
Hagelstein [5]	2D fusion	1–4 keV X-rays
Takahashi [1]	4D fusion	4 keV X-rays, MeV alphas
Kucherov [3]	Photon stimulation of nucleus	Alpha decay, fission
Mayer [6]	Tight molecular binding of light particles	4.7 keV X-rays
Meuhlenberg, Mills, others [2,4]	Collapsed electron states	$\sim$4 keV X-rays

many AHE theories predict the production of low energy X-rays or MeV alpha particles, there has always been a fundamental problem in trying to detect such emissions. This is because low energy radiation would normally be attenuated to undetectable levels by absorption processes acting within the electrolyte, electrodes, and/or calorimeter structures of a standard electrochemical cell. Table 1 summarizes prior theoretical predictions of nuclear emissions expected to emanate from active cathodes which are producing anomalous heat by various postulated processes.

2. Experiment Concept and Design

A series of experiments were performed using custom electrolytic cells with two types of thin membrane cathodes having 0.1 M LiOD electrolyte on one side and air on the other. The first type of cathodic membrane used a 1 μm silicon nitride film coated with 200 nm of platinum (Pt) and palladium (Pd). Three different electrolysis co-deposition methods were used during electrolysis using this type of cathode: D/Pd deposited on Pt cathode from a Pd anode; D/Pd from a Pd salt solution; and D/Pd from a Pd nanoparticle suspension. The second type of cathodic membrane used a 30 μm thick palladium foil. Both of the above cathode types were electrolyzed using constant current for periods of time ranging between 1 and 90 days.

An electrochemical cell was fabricated that permits emissions of low energy X-rays to be detected with very little attenuation by using a thin membrane cathode. (Note: Si_3N_4 membranes are obtained from Silson LTD.) X-ray absorption calculations indicate that for the thin Si_3N_4 membrane, absorption is 90% at 1 keV, and rapidly decreases to 10% at 2 keV. Since a Pd film on the membrane would delaminate when charged with deuterium or hydrogen, 100 or 200 nm Pt film was sputter deposited on the membrane for the anode.

Most calorimeters have sensitivity in the range of 1–10 mW. Suppose that AHE is active much of the time but at micro-, nano-, or picowatt levels. A standard calorimeter would be unable to detect such low levels of power output. If 2 keV X-rays are emitted at $\sim$1 Hz into a background $<$0.2 counts per second, the corresponding heat sensitivity is $<$100 Attowatts. (This sensitivity calculation includes transmission through 100 nm Pt, 100 nm Pd, 1 μm Si_3N_4, 2 mm Air, and measured background X-ray counts.)

Advantages of the above method are as follows.

- Potential sensitivity to detect nuclear emissions corresponding to $<$1 Femtowatt of anomalous heat.
- Ability to directly detect 1–30 keV X-rays.
- Detection of MeV alphas via X-ray induced fluorescence generated from particle impacts on Pd, Pt, and Si cathode structures.
- Enable direct testing for energetic emissions predicted by many different AHE theories including non-nuclear proposals.

Disadvantages of the above method are as follows.

- The novel thin film anode/cathode/electrolyte combinations have not previously been demonstrated to reliably produce excess heat. However, a Pd anode/Pt cathode combination did produce 10% excess at

"Energetics Inc." in 2005 (ref. unpublished internal data) and Pd nanoparticles in electrolyte produced excess heat at SKINR in 2014–2015 (ref. unpublished internal data).

- Pd foil loaded from just one side is not known to yield AHE but similar size and thickness foils certainly can yield anomalous heat when charged from both sides.

Figure 1 is a schematic representation of an electrochemical cell which shows a thin cathodic membrane and the relative location of the detector used to collect X-ray spectral data in recent experiments at SKINR and ENEA.

The X-ray detector used for most of the data collection at SKINR was an Amptek XR100-CR Si PIN diode connected to an Ortec multichannel analyzer running Maestro-32 software. This detector was sensitive to 1–30 keV X-rays and had a spectral resolution of $<$0.3 keV FWHM. Time resolved spectra were collected for 100-second time intervals, compared to nominal ambient background levels using a custom software program, and then archived into a file for future reference. The custom program compared the amplitude of the X-rays detected in each of 4096 energy bins relative to specific threshold values which were previously determined as being the nominal ambient background level expected at that energy. If ANY threshold value was found to be exceeded the program would flag the data set as a *possible* event, write a summary of the specific data which exceeded the threshold into a text file, and generate a real time audible alarm.

The X-ray detector used for data collection at ENEA was an Amptek X123 Si PIN diode containing an integrated multichannel analyzer and having a spectral sensitivity and resolution nearly the same as stated for SKINR's detector. Data collection using the X123 detector at ENEA was not time resolved but instead was time integrated over the entire data collection period.

3. Experimental Results at SKINR

Figure 2 shows a calibration spectrum of the X-ray detector which used a ^{241}Am radioactive source while Fig. 3 is a typical background spectrum collected by acquiring data for 200,000 s. The ambient background was found to be very repeatable and essentially free of any naturally occurring radioactive source emission lines. Figure 4 shows two 100-second interval runs, one using a 30 μm thick Pd cathode and the other using a PdCl co-deposition of Pd on a

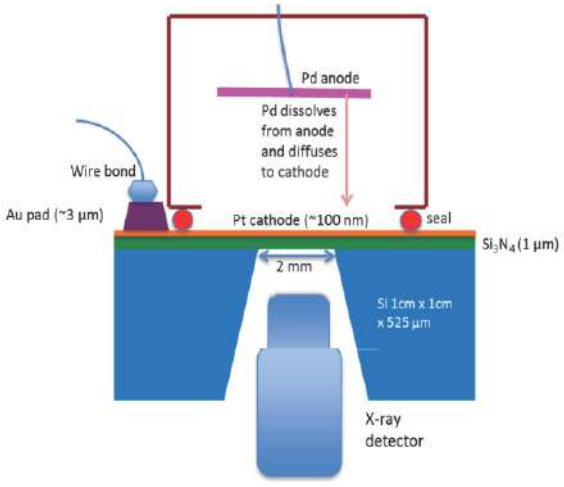

Figure 1. The cathodic membrane electrolytic cell used in SKINR and ENEA X-ray spectra tests.

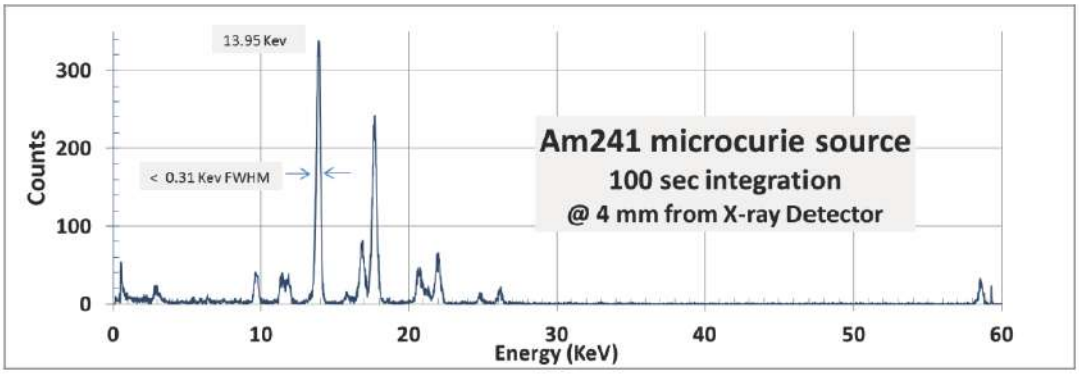

Figure 2. SKINR X-ray spectral calibration data acquired from an ^{241}Am microcurie test source.

SiN membrane. During each of the experimental test runs the custom search program was set to automatically trigger manual inspection if any channel exceeded five counts. At the end of each data run an even more sensitive check was performed by adding together all of the individual time-resolved test spectra and searching for the presence of any weak X-ray lines. Note that the above experimental technique would have also allowed any MeV alphas, if present, to have been detected by impact induced fluorescent X-rays from within the experimental setup materials including Pd $\sim$21.2, 23.8 keV $K_{\alpha,\beta}$; Pt $\sim$9.4, 11.1 keV $L_{\alpha,\beta}$; Pd $\sim$2.8, 3.0 keV $L_{\alpha,\beta}$; or Si$\sim$1.7,1.8 keV $K_{\alpha,\beta}$.

Table 2 contains a summary of the membrane characteristics and electrolytic cell parameters used in the various X-ray spectral data test runs at SKINR. For most of these data sets continuous X-ray data was collected and stored at 100 s intervals throughout the listed duration of the test so as to create a time-resolved view of each experiments X-ray spectral emissions. A re-occurring computer error (now fixed) caused some experiments to miss spectral data during portions of the total test run time. Even in those cases, however, there were many, many, 100 s long X-ray spectral data sets which were successfully acquired, stored, and reviewed.

Table 2. Membrane electrolytic cell parameters used in SKINR X-ray spectra tests.

Cathode		Anode	Electrolyte	Current density (mA/cm^2)	X-ray detector model/cell	Duration (days)
Membrane	Coated					
Si_3N_4	100 nm Pt	Pt	$PdCl_2$+LiCl	0.01–0.1	XR100/Glass cell	10
Si_3N_4	100 nm Pt	Pd	0.1 M LiOD	0.2–1	XR100/Glass cell	5
Si_3N_4	100 nm Au	Pd	0.1 M LiOD	0.5–2.5	XR100/Glass cell	11
Si_3N_4	100 nm Pt	Pd	0.1M LiOD	0.25–2	XR100/Glass cell	8
Si_3N_4	100 nm Pt	Pd	0.1 M LiOD	0.25–6	XR100/Glass cell	60
Si_3N_4	100 nm Pt	Pt	PdNp+0.1 M LiOD	0.1–6	XR100/Glass cell	7
Si_3N_4	100 nm Pt	Pt	PdNp+0.1 M LiOD	0.1–6	XR100/Glass cell	12
Si_3N_4	100 nmPd	Pt	PdNp+0.1 M LiOD	0.1–6	XR100/Glass cell	1
Si_3N_4	100 nm Pt	Pt	PdNp+0.1 M LiOD	0.1–6	X123/Glass cell	20
Si_3N_4	100 nm Pt	SS	PdNp+0.1 M LiOD	0.1–6	X123/Glass cell	90
Foil	Pd 30 μm	Pt	0.1 M LiOD	0.25–6	XR100/ Peek cell	30
Foil	200 nm Au on 25 μm Pd	Pt	0.1 M LiOD	0.5–10	XR100/ Peek cell	20

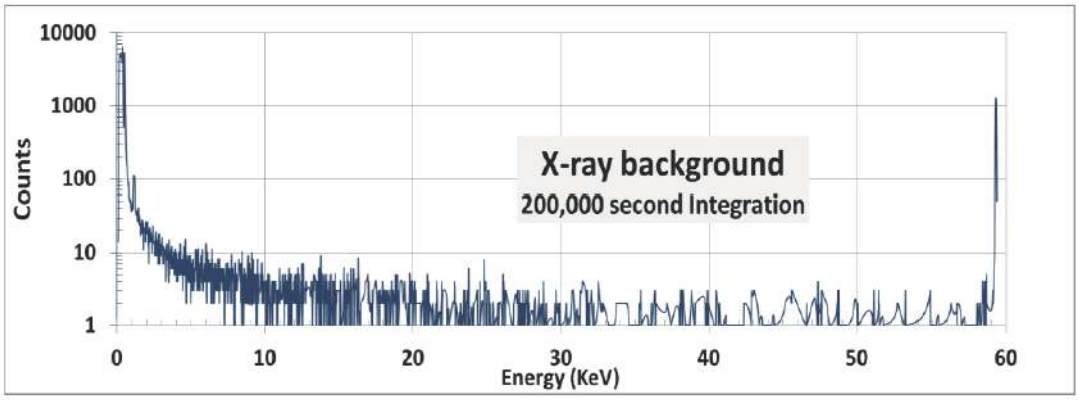

Figure 3. SKINR typical X-ray ambient background spectral data acquired and integrated for 200,000 s.

4. Experimental Results at ENEA

Additional tests using a similar cathodic membrane electrolytic cell and X-ray detector were independently conducted by ENEA, Italy. Figure 5 contains two representative spectrums from one of ENEA's experiments. One spectrum (blue trace) shows the nominal ambient background and has been scaled to match the same data acquisition period as was used in the collection of the active cell membrane data (red trace). This test data used a 25 μm thick Pd foil coated with 100 nm Au on the air/detector side and the plot shows X-ray spectra from 1 to 32 keV that was continuously accumulated for 5.8 days. The background and active cell spectra closely match one another at all wavelengths and are featureless except for an obvious line near 22 keV. The source of this line has been conclusively determined to be due to Pd $K_{\alpha,\beta}$ X-rays at 21.1 and 23.8 keV and it is created by fluorescence from 100 keV gamma rays emitted by a Bi radioactive isotope contaminant within the Pb shielding.

Table 3 contains a summary of the membrane characteristics and electrolytic cell parameters used in the various X-ray spectral data test runs at ENEA. For each of these runs a single time integrated X-ray spectrum was acquired

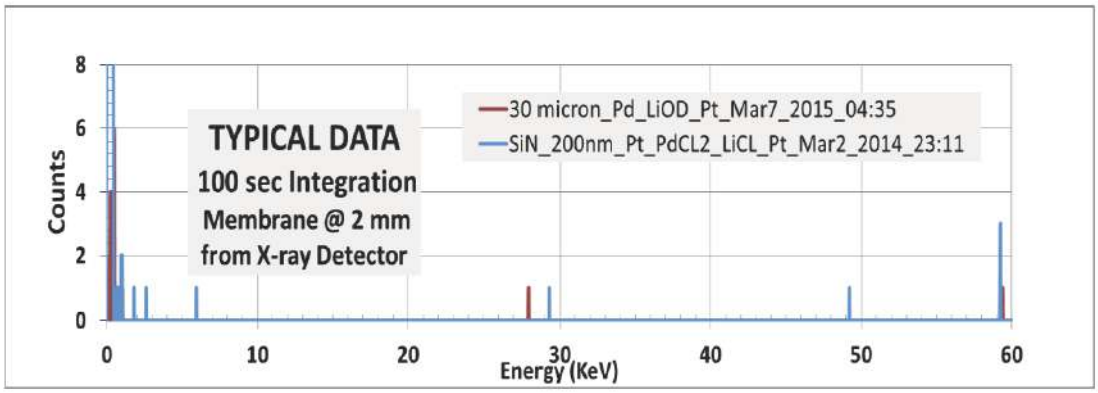

Figure 4. SKINR typical X-ray spectral data acquired from either a co-deposited Si_3N_4-Pd membrane or a thin Pd foil electrolytic cell.

Table 3. Membrane electrolytic cell test parameters used in ENEA X-ray spectra tests.

Cathode		Anode	Electrolyte	X-ray detector model	Duration (days)
Membrane	Coated				
Foil	100 nm Pt	Pd	0.1 M LiOD	Amptek X123	0.71
Foil	100 nm Au	Pd	0.1 M LiOD	Amptek X123	0.82
Foil	200 nm Au on 25 μm Pd	Pd	0.1 M LiOD	Amptek X123	5.8

which spanned the listed experimental duration.

5. Conclusions

It was hoped that these membrane X-ray experiments would reveal important information to help discriminate between the many different theoretical hypotheses proposed to explain the physical mechanisms which underlie and create anomalous heat effect (AHE) in electrolytic cells. No X-rays, however, were detected at SKINR above ambient background levels during several months of experimental runs. While ENEA data initially showed a couple X-ray peaks, these were later conclusively demonstrated to be due to Pd $k_{\alpha,\beta}$ fluorescence induced by 100 keV gamma rays emitted from Bi contaminants within external Pb shielding. While these detections showed that the experiment was definitely sensitive to fluorescence induced Pd X-rays the final result from ENEA was the same as from SKINR: *No X-rays were detected in any experimental runs.* The above null results at ENEA and SKINR imply the following derived conclusion.

If AHE involves the emission of low energy X-rays and/or MeV alpha particles then there are only two possibilities. One is that AHE was not active in any of the test runs at power levels between ∼1 Femtowatt and ∼1 mW and was therefore in an OFF state. The other possibility is that the cathode was actually in an ON state but was producing both imperceptible low levels of anomalous heat and undetectable levels of low energy radiation. In the latter case, our near individual particle sensitivity to radiation yields the conclusion that NONE of the theories in the table are likely to be viable.

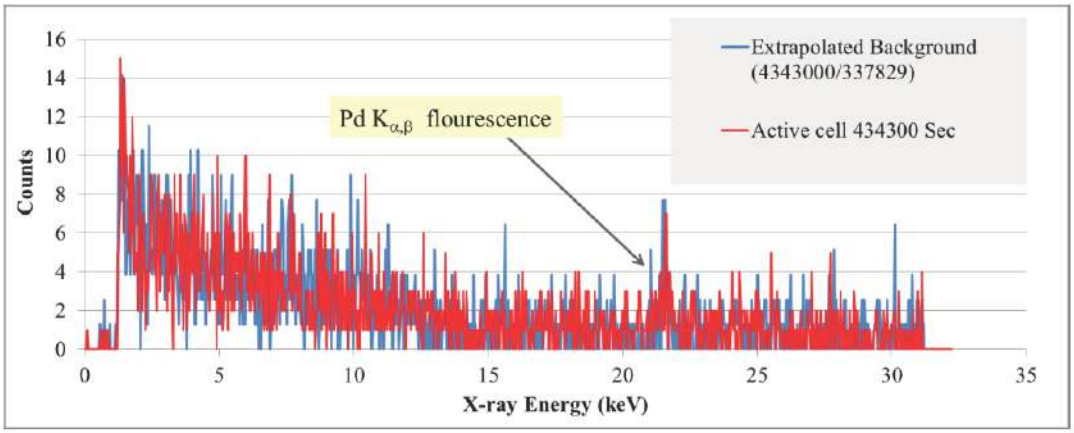

Figure 5. ENEA acquired X-ray spectra from a membrane electrolytic cell compared to an ambient background spectra scaled in intensity to match the same data acquisition time.

It was also concluded from the above experiments and their null X-ray results that the continuation and improvement of membrane electrolytic cell testing is warranted. While the fragile Si_3N_4-Pd co-deposited membranes only survived from 1 day to 3 months, the solid Pd cathode foils seemed to have an unlimited lifetime and could therefore be run for the longer times previously needed to produce substantial levels of anomalous heat. Several useful additions to the membrane experimental setup were also identified. These included placement of the membrane system in a calorimeter, searching for RF emission (RF was first detected by NRL [7] and later confirmed by ENEA [8], and/or stimulating the cathode with RF fields, acoustic waves, electrical pulses, or lasers.

Acknowledgement

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