ELECTROLYSIS OF D_2O WITH TITANIUM CATHODES: ENHANCEMENT OF EXCESS HEAT AND FURTHER EVIDENCE OF POSSIBLE TRANSMUTATION

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Abstract:
Using Ti-Pt electrodes in closed electrolytic cells containing D_2O-H_2SO_4 electrolyte, evidence for excess thermal power generation has been observed (i.e. Power out – Power in > 0). It had been noted that experiments (8-cell) with smaller cathodes (larger perimeter to area ratio) performed better than experiments (SEC system) in which larger cathodes (smaller perimeter to area ratio) were used. In an effort to increase the magnitude of the excess power output, slits were introduced into the larger cathodes to increase the perimeter to area ratio. Two SEC systems were used during the course of these experiments. Using data from the first SEC system we find that four of seven (57%) of the experiments with slit cathodes showed an excess thermal power, averaging 322 mW (ranging from 136 to 509 mW) and five of fourteen (36%) of the experiments with no cathode slits gave excess thermal power, averaging 171 mW (ranging from 115 to 233 mW). Overall, 10 of 13 (77%) of the experiments with slit cathodes showed excess power while only 5 of 15 (30%) of the experiments with no slits in the cathodes showed excess power. This result shows an increase in both the magnitude and reproducibility of the excess power output effect. In addition, Neutron Activation Analysis (NAA) was performed on several cathodes (post-experiment) where greater concentrations of unexpected elements are found in those cells that showed excess power compared to those cells that did not show excess power.

Introduction:
Previously we have shown a method of increasing both the reproducibility and magnitude of the excess power generated by optimizing the reduction in thickness of the cathode by cold rolling[1]. Recent evidence has been found that may indicate another important variable that could be used to increase the magnitude and repeatability of excess thermal power production in electrolytic cells containing Ti/Pt electrodes in D_2O/H_2SO_4 electrolyte.

Two calorimeter systems, an eight-cell, small cathode system, and a single cell Seebeck Envelope Calorimeter (SEC) system with a much larger cathode, are used in this research[1]. These systems measure the thermal power output of closed electrolytic cells employing recombination catalyst and containing Pt anodes and Ti cathodes in D_2O-H_2SO_4 electrolyte. The Ti cathodes in the eight-cell experiments typically have surface area of about 23 mm^2, perimeter of about 10 mm, and current density of about 2 A/cm^2. For those cells that showed excess power, the average excess power generation was 232 +/- 20 mW, which represents an increase of about 13% over the power input (or 113% power out/power in). Excess power output increased with increased current density. The highest power output observed was 125% of the power input. An effort was made to increase excess power by increasing the size of the cathode to about 400 mm^2 in the SEC experiments. However, the larger cathodes did not appear to increase the excess power output. The smaller cathodes have larger perimeter / area ratios than the larger cathodes. In an attempt to understand this lack of increased output with increased area, we then investigated the relationship between the perimeter / area ratio of the cathode and excess heat output over a series of 28 SEC experiments.

Methods and Materials:
Two Seebeck Envelope Calorimeters (SEC) from Thermionics Corp. were employed during these experiments. Each SEC has an inner cavity measurement of 18 cm by 18 cm by 18 cm. Heat flows from the experimental cell, positioned on a raised platform in the center of the SEC cavity, through the surrounding air inside the calorimeter and through the SEC envelope, consisting of 16 thermocouples per cm^2, to an aluminum jacket that incorporates constant temperature water-cooling coils. If the inner cavity has a higher temperature than the water jacket, the resulting heat flux produces a positive electrical signal output from the SEC in the 0-100 millivolt range over the span of power levels used in these trials.

The first SEC was calibrated using 33 data points from Pt-Pt control cells with H_2O-H_2SO_4 electrolyte. The SEC gives a linear input vs. output relationship over the range of interest:

$$\textit{Power Output (Watts)} = 0.202 \pm 1.4\times10^{-6}\ \textit{W/mV} \times \textit{SEC Output (mV)} + 0.045 \pm 0.008\ \textit{W} \quad (1)$$

and has a coefficient of determination R^2 of 0.9999. The intercept in the slope of the line equation (1) reflects an adjustment of 0.045 W. Thermal leakage along wires fed into the SEC via a 0.6 cm tube inserted into the inner cavity of the SEC accounts for this adjustment. These wires include the power leads to the electrolytic cell, voltage measurement wires and six thermocouple wires used to measure the temperature of the outside surface of the cell. Because of uncertainties inherent in the Pt-Pt control cell such as recombination and evaporative weight loss, we do not claim excess power production unless then output exceeds 100mW.

The second SEC reduces these uncertainties by utilizing a new SEC calorimeter and a resistive cell instead of a Pt-Pt cell. There are no mass losses from the resistor cell, but there are losses in the Pt-Pt cell. Therefore, the excess power calculation will be conservative using the resistive cell. In addition, we no longer employ the calibration curve but instead perform a direct comparison of the power in / signal outputs of the resistive control immediately after the experimental run to the power in / signal outputs for the experimental cell. Since this calibration is more accurate than the older (first) method (a Pt-Pt control cell) we will now claim excess power is generated if the excess power output exceeds 3σ using the second SEC, Table 3.

The Ti foil used in this study (99.99%) was obtained from two suppliers: Aldrich Chemical Co. and Alfa Aesar. Aldrich Chemical Company reported impurity levels in batch 1 (Lot: 01230AQ) of: 30 ppm Ca and 8 ppm Al whereas batch 2 (Lot: 04624KI) was found to contain: 3.8 ppm K, 3.2 ppm Al, 2.7 ppm Fe, 0.9 ppm Ca, 0.5 ppm Li and 0.3 ppm Na. The foil obtained by Alfa Aesar (Stock: 43676, Lot: E25K24) reported impurities of 31 ppm Al, 76 ppm C, 29 ppm N, and 70 other elements with concentrations <10 ppm including Pt < 0.05 ppm and Au < 0.05 ppm. The Ti foil from Aldrich batch 1 was used in SEC 2, 3, 4, 5, 6, 7, 8, 9, 10, 11. Aldrich Ti batch 2 was used in SEC 18, 19, 20, 21, 22, 23 and Alfa Aesar Ti foil was used in SEC 13 and 17. Each of the cathodes used in this paper were cold rolled 20% in addition to any cold rolling performed by the supplier.

Results:

To increase the perimeter / area ratio, vertical slits were cut into the Ti cathodes of thirteen SEC cells. Six of the Ti cathodes were cut using stainless steel surgical scissors and seven Ti cathodes (SEC 9, for example) were cut using an Electrical Discharge Machining (EDM) process (discussed in more detail later).

Introducing these slits increased the perimeter / area ratio to an average value of about 0.5 mm^{-1}, which was twice that of the SEC cathodes without slits (0.25 mm^{-1}). The current density was lower for most of the SEC experiments that used cathodes with slits, about 0.56 A/cm^2, compared with 1.01 A/cm^2 for the cathodes without slits. A summary of results of cathodes with slits can be found in Table 1, experiments without slits is found in Table 2 and the remaining experiments that utilized the second SEC system is found in Table 3.

Table 1: Summary of cells in which slits were cut into the cathodes using the first SEC system.

Slits in Cathodes	Excess Power (Watts)	Power In (Watts)	Current Density (A/cm^2)	Exp. Runtime (Min)	Excess Heat Time (Min)	Mass Loss (g/Sec)	Power to Add (W)	Area (mm^2)	Perimeter /Area Ratio (mm^{-1})
Excess									
SEC 6 Before	0.281 ± 0.007	10.37	0.56	1155	461	$1.1x10^{-5}$	0.166	356	0.20
SEC 6 After	0.322 ± 0.011	11.54	0.53	7467	3748	$5.9x10^{-6}$	0.087	379	0.34
SEC 7	0.136 ± 0.009	10.71	0.71	8750	5721	$4.1x10^{-5}$	0.609	281	0.41
SEC 8	0.509 ± 0.017	16.67	0.45	13331	10503	$2.6x10^{-6}$	0.038	553	0.37
SEC 9	0.370 ± 0.007	12.27	0.31	5184	2764	$9.2x10^{-7}$	0.018	651	0.62

Table 2: Summary of cells using the first SEC system where no slits were cut into the cathodes.

No Slits in Cathodes	Excess Power (Watts)	Power In (Watts)	Current Density (A/cm^2)	Exp. Runtime (Min)	Excess Heat Time (Min)	Mass Loss (g/Sec)	Power to Add (W)	Area (mm^2)	Perimeter /Area Ratio (mm^{-1})
Excess									
SEC 2	0.115 ± 0.007	12.05	0.65	9455	7132	$4.4x10^{-5}$	0.656	310	0.19
SEC 3	0.217 ± 0.018	9.20	0.59	14481	12939	$4.1x10^{-6}$	0.061	340	0.15
SEC 18	0.132 ± 0.011	8.46	0.44	10155	6039	$7.3x10^{-6}$	0.109	365	0.21
SEC 19	0.233 ± 0.011	9.19	0.51	13590	10499	$4.9x10^{-6}$	0.073	394	0.20
SEC 21	0.119 ± 0.011	8.25	0.50	10130	6242	$8.3x10^{-6}$	0.123	441	0.17

While reviewing Tables 1 and 2, keep in mind that the error for the SEC has an average value of ± 11 mW. Nonetheless, we do not claim excess power until the observed excess power output exceeds 100 mW.

Table 3: Experiments performed using the second SEC system.

New SEC Calorimeter	Max Excess Power (Watts)	Power In (Watts)	Current Density (A/cm^2)	Exp. Runtime (Min)	Excess Heat Time (Min)	Mass Loss (g/Sec)	Power to Add (W)	Area (mm^2)	Perimeter /Area Ratio (mm^{-1})
Cathodes with Slits									
SEC 27Before	0.019 ± 0.014	11.00	0.42	2804	553	$3.0x10^{-5}$	0.437	454	0.17
SEC 27 After	0.052 ± 0.014	10.69	0.40	4288	3253	$5.0x10^{-6}$	0.074	527	0.64
SEC 29	0.048 ± 0.010	12.66	0.44	5760	1423	$4.6x10^{-6}$	0.073	551	0.67
SEC 30	0.076 ± 0.009	12.78	0.46	8399	2590	$2.9X10^{-6}$	0.047	544	0.64
SEC 31	0.051 ± 0.008	12.60	0.76	1539	367	$5.4X10^{-6}$	0.096	332	0.58
SEC 32	0.058 ± 0.007	15.73	0.87	1440	112	$9.8X10^{-6}$	0.158	287	0.65
SEC 33	0.098 ± 0.013	10.89	1.01	1363	185	$1.6X10^{-6}$	0.023	223	0.64

The first indication that the perimeter / area ratio might be important occurred during the SEC 6 experiment. With a power input of 10.37 W and a current density of 0.53 A/cm^2, the excess power output was steady at 281 mW for about 4 hours. The experiment was then halted, the cathode was removed from the experimental cell, four slits were cut into the cathode using surgical scissors and then the cathode was placed back into the cell for further operation. At first, the excess power output decreased to 162 mW with a power input of 10.15 W and a current density of 0.53 A/cm^2. After about 43 hours with power output climbing, the excess power output leveled off at 322 mW and remained steady for 40 hours with a power input of 11.54 W and a current density of 0.53 A/cm^2. SEC 27, Table 3, also shows an increase in excess power output ranging from no excess power output (19mW± 14mW) before modification to 52mW ± 16mW excess power after slits were introduced using the EDM process (see below for details).

Each of the cathodes in Table 1 contained slits, which were cut using surgical scissors except for SEC 9. The slits in the SEC 9 cathode were cut using an Electrical Discharge Machining process (EDM) by Wire EDM Specialties Co. in Clackamas Oregon, USA. This process heats a narrow section (~1μm) of the cathode to a point above the vaporization temperature of Ti. The sample is also bathed in a coolant during the EDM process. The high precision of the EDM process allowed for 21 slits to be cut into the cathode (13 slits are considered to be exceptional number of slits using surgical scissors). Before the EDM process, SEC 9 had a perimeter of 67 mm and an area of 331 mm^2. After the EDM process, the perimeter was found to be 401 mm (an increase of ~ 600%) and the area was 651 mm^2 (an increase of ~ 200%). The final perimeter / area ratio of 0.62 mm^{-1} was three times that of the beginning (pre-slit) ratio of 0.20 mm^{-1}. SEC 9 had the largest perimeter / area ratio and the smallest current density of all of the experiments reviewed in this paper and returned the second highest excess power output of 370 mW (with a power input of 12.27 W). Table 3 shows the results of the seven experiments performed using the second SEC system. The EDM process was also employed in each of the experiments with slits found in Table 3.

Neutron activation analysis (NAA), a bulk analysis technique, was performed after electrolysis on both the electrolyte and the cathodes of several SEC and eight-cell experiments. The first group of SEC experiments analyzed had cathodes cut from Aldrich, lot # 01230AQ, 99.99% pure Ti foil. The results for these analyses are shown in Table 4. The second group of SEC experiments analyzed were cut from Aldrich batch 1 and batch 2 (SEC 6, 17, 19, 20, 26). The second NAA group analyzed showed null results for SEC 17, 19 and 20. The results for SEC 6 and 26 are lower than blanks run at the same time. Therefore, with no beneficial information available, the data for these analyses will not be displayed here for space considerations.

Table 4: Results of NAA analysis on SEC experiments.

	SEC 4	SEC 7	SEC 8	SEC 11
Excess Power (W)	- 0.011 ± 0.010	+ 0.136 ± 0.010	+ 0.509 ± 0.007	+ 0.016 ± 0.011
Pt-197 on Cathode (ppm)	$3.53x10^{3}$ ± $6.66x10^{2}$	$4.40x10^{2}$ ± $1.41x10^{2}$	0.00 ± 0.00	$5.33x10^{3}$ ±$1.01x10^{3}$
Au-197 in Electrolyte (ppm)	$3.76x10^{-6}$ ± $8.29x10^{-6}$	$2.02x10^{-5}$ ± $5.71x10^{-6}$	$1.48x10^{-5}$ ± $4.76x10^{-6}$	$1.14x10^{-5}$ ± $4.61x10^{-6}$

Statistically significant amounts of platinum were found in larger amounts in the cathodes that did not show excess heat (SEC 4 and SEC 11) compared to the cathodes that did show excess heat (SEC 7 and SEC 8). A plausible explanation for the appearance of Pt on the cathode might be that the NAA analysis picked up Pt from the Pt (lead wire) / Ti spot weld interface. The interface junction was present on the cathodes of SEC 4, SEC 7 and SEC 11 cathodes but is not present in the SEC 8 cathode.

Statistically significant amounts of gold were detected in the electrolyte from the two excess heat producing experimental electrolytes SEC 7 (+ 0.136 ± 0.010 W) and SEC 8 (+ 0.509 ± 0.007 W) but no statistically significant Au was found in the electrolyte of the non-excess heat producing experiments (SEC 4 and SEC 11). We previously reported the detection of localized concentrations of Au by surface analysis of Pd cathodes from cells that produced excess heat [2]. The Au could result from the transmutation of Pt from the anode or lead wires in both the Pd and the Ti cathode cells.

Gold has also been found in two of the 8-Cell experimental cathodes/ electrolyte as well. The results are shown below in Table 5.

Table 5: NAA results of final two 8-cell experimental cathodes and electrolyte.

	JWMS 3 Cathode	JWMS 3 Liquid	JWMS 4 Cathode	JWMS 4 Liquid	Blank 1	Blank 2
Excess Power (W)	+0.210 ±0.030		+0.070 ±0.043			
Au-197 Detected (ppm)	9.69×10^{-2} $\pm1.53\times10^{-2}$	1.15×10^{-4} $\pm2.53\times10^{-5}$	2.48×10^{-2} $\pm4.31\times10^{-3}$	5.70×10^{-5} $\pm1.51\times10^{-5}$		
Au-197 Detected (g)	1.69×10^{-9} $\pm2.58\times10^{-10}$	4.44×10^{-10} $\pm9.77\times10^{-11}$	4.41×10^{-10} $\pm7.60\times10^{-11}$	2.16×10^{-10} $\pm5.70\times10^{-11}$	2.75×10^{-10} $\pm5.23\times10^{-11}$	8.92×10^{-11} $\pm2.26\times10^{-11}$

The NAA results include the outcome of blanks that were also run during the NAA analysis. Each of the blanks showed the presence of Au. Blank #1 showed a mass concentration of $2.75\times10^{-10} \pm 5.23\times10^{-11}$ grams Au. This negates the JWMS 4 Liquid result in Table 5. The data in Table 5 indicates that higher concentrations of Au were found in the cathode and electrolyte of JWMS 3, an excess power-producing cell, than was found in the analysis of JWMS 4, a non-excess power-producing cell. JWMS3 had about three times the excess power production and about four times as much Au found on the cathode. This result indicates that the hypothesized transmutation of Pt to Au could be indicative of a nuclear process that increases with increasing excess power but does not greatly contribute to the excess thermal power output.

Discussion and Conclusion:

Overall, 10 of 13 (77%) of the experiments with slit cathodes showed excess thermal power while only 5 of 15 (30%) of the experiments with no slits in the cathodes showed excess thermal power. Using the data from the first SEC alone shows that four of seven (57%) of the experiments with slit cathodes showed excess thermal power, averaging 322 mW (ranging from 136 to 509 mW) and five of fourteen (36%) of the experiments with no cathode slits gave excess thermal power, averaging 171 mW (ranging from 115 to 233 mW). Thus, an increase in the perimeter / area ratio of the cathode appears to increase both the reproducibility and the magnitude of excess power production in Ti-Pt electrolytic cells containing an electrolyte of D_2O-H_2SO_4.

In addition, the data presented for 12 of the 22 total NAA analysis (11 cathodes and 11 electrolyte samples) seems to indicate that transmutation products occur in higher atomic concentrations in those cells that showed evidence for excess power generation.

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References:

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