Twenty Year Review of Isoperibolic Calorimetric Measurements of the Fleischmann-Pons Effect

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Introduction: Why Choose a Dewar Isoperibolic Calorimeter?

The Dewar type of isoperibolic calorimeters developed by Fleischmann and Pons provides a wide dynamic range for both the cell temperature and cell input power. The experimental temperature range can be as great as the liquid range of the electrolyte system (3.82°C to 101.42°C for pure D_2O). For typical D_2O+0.1 M LiOD electrolytes, the temperature range used is typically from 20°C dictated by the bath temperature up to the boiling point of the electrolyte. The maximum cell input power will vary with the size of the cell, but input powers up to 10 W are possible before the cell contents are driven to boiling temperatures.

The ability provided by the Dewar type cells for directly observing processes inside the cell is a very important advantage. This is especially important for co-deposition and cell boil-off experiments. Other calorimetric systems generally do not provide for visual observations inside the cell.

The relatively low cost of the Dewar type cells is another important factor for the selection of this isoperibolic system. This makes possible the simultaneous use of multiple cells involving different experiments in a single water bath of sufficient size for handling the total heat output. The major cost is simply the construction of glass Dewar cells with a high vacuum between the inner and outer glass surfaces. The top portion (about 30%) needs to be silvered to minimize the effect of the decreasing electrolyte level caused by the electrolysis of D_2O to produce D_2 and O_2 gases. Larger cells will obviously exhibit smaller changes in the electrolyte level during electrolysis.

Another important positive feature of these Dewar cells, as well as other open systems, is that they are self-purifying. Ordinary H_2O will act as a poison to the Fleischmann-Pons effect (FPE) in D_2O+0.1 M LiOD electrolytes. Because H is preferentially electrolyzed versus D at the cathode, any H_2O contamination will be gradually removed during electrolysis. In closed systems, the initial H_2O contamination will remain trapped throughout the experiment. Not to be overlooked is the inherent safety of an open system that allows the deuterium and oxygen gases to escape as fast as they are generated.

The main heat transfer pathway for Dewar isoperibolic cells is via electromagnetic radiation (mostly infrared radiation). The radiative heat transfer coefficient, therefore, can be estimated by using the Stefan-Boltzmann constant and the experimental surface area of the inner, unsilvered portion of the glass surface of the Dewar cell. This provides a useful guide to the integrity of the Dewar vacuum and the extent of minimization of heat transfer pathways via conduction. Because of the predominate heat transfer by electromagnetic radiation, this Dewar cell system has no memory effect. For example, gas bubbles or stagnant layers that form on the inner cell wall will exert no effect on the radiative heat transfer coefficient. Any media in the cell will suffice that maintains the temperature of the inner cell wall.

The Fleischmann-Pons Dewar System

The Fleischmann-Pons (F-P) isoperibolic calorimetry using a Dewar type cell evolved through various designs dating back to the early 1980's. The major changes involved the dimensions selected for the cell as well as incorporating the silvering of the top portion of the cell [1,2]. Proper scaling of the system is critical because the cell diameter and length determine the volume of electrolyte used, the rate of change of the electrolyte level, the effectiveness of stirring by the electrolysis gases, the dynamic range for power input, and the magnitude of the calorimetric constants. These changes led to the ICARUS (Isoperibolic Calorimetry: Acquisition, Research and Utilities System) series of cells used in later experiments.

The ICARUS 1 to 3 calorimeters were for lower temperatures up to boiling, and the ICARUS 4 to 9 calorimeters allowed long-term maintenance of boiling conditions. Results of ICARUS 9 experiments have been presented [3]. The ICARUS 10 to 13 were designed and constructed for further studies of boiling, but they were never put in use [4]. A schematic diagram for the ICARUS-14 calorimeter is available, but this calorimeter was never constructed[4]. An example of the ICARUS-1 type cell used at the New Hydrogen Energy (NHE) laboratory at the Sapporo, Japan from 1994 to 1998 had a filled electrolyte volume of 90 cm^3, an inner diameter of 2.5 cm, an outer diameter of 4.2 cm, and a length of 25.0 cm with the top 8.0 cm silvered. A similar Fleischmann-Pons Dewar cell was used in France by Longchampt and Bonnetain [5].

The modeling and mathematical equation of the Fleischmann-Pons Dewar isoperibolic calorimeter have been previously presented [1,2,4,6,7]. A detailed version of this present paper containing all the equations is available electronically [8]. The correct equations for modeling isoperibolic calorimetry using open cells are now well established and can give highly accurate results [2,8]. There has been no challenge to these calorimetric equations in any refereed scientific publication even after nearly twenty years. Therefore any institution that performs accurate isoperibolic calorimetry using open cells must correctly account for all of the following power terms:

1. Power for the calorimetric system itself due to changes in the cell temperature.
2. Electrical power due to the electrochemical reactions.
3. Power due to the transfer of heat by electromagnetic radiation.
4. Power due to the transfer of heat by conduction.
5. Power carried out of the open system by the gases exiting the cell including D_2O vapor.
6. Power applied to any internal heater.
7. Power due to the rate of any pressure-volume work done by the electrogenerated gases.
8. Power due to any anomalous source (Excess Power).

Isoperibolic calorimetric measurement in 1989 at three influential institutions prematurely squashed the public and scientific considerations of the Fleischmann-Pons (F-P) Effect. These three institutions (Caltech, M.I.T., Harwell) did not account for many of the power terms listed above.

California Institute of Technology Calorimetry

The initial publication by Caltech on the F-P effect was received by Nature on 23 May 1989 [9]. This means that all work was completed in less than two months after the initial announcement of the F-P effect (March 23, 1989). It is remarkable that Caltech claimed completion of multiple calorimetric experiments in this short time span because completion of a single F-P experiment generally requires 4 to 8 weeks or more of electrolysis.

The first step in evaluating this Caltech calorimetry is to look for the required power terms in the modeling equations. The Caltech paper is almost devoid of equations for these power terms. Only terms for the electrochemical power and an expression for the total power could be identified [8]. The Caltech calorimetric cell consisted simply of a Dewar flask containing 30 mL of electrolyte, but the Dewar walls contained 1 atm of air, hence there was no vacuum and heat conduction would dominate over electromagnetic radiation. The most disturbing aspect of the Caltech report concerns the heating coefficient that was allowed to change with time to give zero excess power. This heating coefficient should have a nearly constant value as found in their control study using ordinary water. The Caltech heavy water experiment is more correctly interpreted as an excess power effect [8].

Massachusetts Institute of Technology Calorimetry

The two M.I.T. calorimeters used glass wool thermal insulation in the compartment between the cell and water bath [10], thus their main heat transfer pathway was by conduction rather than by electromagnetic radiation as in the F-P Dewar cells. The sensitivity of the Phase II M.I.T. calorimeters was stated as ±40 mW [10] contrasted to ±0.1 mW sensitivity for the F-P Dewar calorimeters [1,2]. From the dimensions of the M.I.T. Pd rod cathodes (0.1 × 9 cm), the expected excess power of about 70 mW would not easily be distinguished from the large calorimetric error reported. The M.I.T. laboratory reported their key calorimetric measurements over a rather short time period (100 hours). The observation of excess power with Pd cathodes in heavy water generally requires 6 days or more of electrolysis.

To M.I.T.'s credit, most of the required power terms were considered in their analysis. The main problem with the M.I.T. experiments is that there is a serious discrepancy between the unpublished raw data showing a small excess power effect and the final published data for their heavy water cell [8]. The data points appear to have been arbitrarily shifted down to make the excess power vanish [8]. Because a "wake" to ridicule cold fusion at M.I.T. was already planned before the experiments were completed, it would have required unusual honestly for M.I.T. to have correctly reported a small excess power effect.

The Harwell Calorimetry

The Harwell (U.K.) laboratory investigated the F-P effect using Dewar calorimetric cells during the early stage (1989) of the cold fusion controversy [11]. These Harwell experiments were hastily performed, and the reported calorimetric error of ±15% falls far short of the ±0.01% error reported by Fleischmann [1,2]. The Harwell publication [11] provides only fragmental information about the power terms and equations used in their calculations for their Dewar cell. Most of the required power terms are missing in their analysis of the data [8]. To the credit of the Harwell group, their raw experimental data was made available to other groups. Several independent analyses of this raw data have reported that excess enthalpy generation was in fact observed in the Harwell study contrary to the conclusions reached by the authors [8].

Summary

The correct equations for modeling isoperibolic calorimetry using open cells are now well established and can give highly accurate results. These calorimetric equations were used to evaluate the Caltech, M.I.T., and Harwell calorimetry performed in 1989. It appears that scientific objectivity was sacrificed by these three influential institutions in order to obtain their desired result of no anomalous excess power effects.

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