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Calorimetric Analysis of a Heavy Water Electrolysis Experiment Using a Pd-B Alloy Cathode

M.H. MILES

*Chemistry and Materials Division, Research Department
Naval Air Warfare Center Weapons Division
China Lake, CA*

M. FLEISCHMANN

*Bury Lodge, Duck Street
Tisbury, Salisbury, Wiltshire SP3 6LJ
England*

M.A. IMAM

*Physical Metallurgy Branch
Materials Science and Technology Division*

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13. ABSTRACT (Maximum 200 words) This study involves the palladium-boron alloy materials prepared at the Naval Research Laboratory (NRL). This new material was developed as part of a collaborative program with NRL and the Naval Air Warfare Center Weapons Division (NAWCWD), China Lake, that was funded by the Office of Naval Research (ONR). Studies at NAWCWD showed that the best reproducibility for excess power was obtained using the palladium-boron materials. The experimental studies described in this report were conducted by Dr. Melvin H. Miles at the New Hydrogen Energy (NHE) laboratory in Sapporo, Japan. Dr. Miles received a six month appointment as a Guest Researcher sponsored by the New Energy Development Organization (NEDO) of Japan. This experiment was conducted in a special Dewar-type calorimetric cell silvered at the top that was developed by Drs. Fleischmann and Pons. The detailed analysis of the experimental data presented in this report was conducted by Dr. Martin Fleischmann. An independent method of data analysis developed by Dr. Miles while he was in Japan was presented in his NEDO Final Report and shows similar trends for the excess heat effect.

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FOREWORD

This study involves the palladium-boron alloy materials prepared at the Naval Research Laboratory (NRL) by Dr. M. Ashraf Imam (see NRL/MR/6170-96-7803, January 9, 1996). This new material was developed as part of a collaborative program with NRL and the Naval Air Warfare Center Weapons Division (NAWCWD), China Lake, that was funded by the Office of Naval Research (ONR). Studies at NAWCWD showed that the best reproducibility for excess power was obtained using the palladium-boron materials supplied by NRL (see NAWCWPNS TP 8302, September 1996). The new experimental studies described in this report were conducted by Dr. Melvin H. Miles at the New Hydrogen Energy (NHE) laboratory in Sapporo, Japan. Dr. Melvin H. Miles received a six month appointment as a Guest Researcher sponsored by the New Energy Development Organization (NEDO) of Japan. Dr. Melvin H. Miles expresses his appreciation to Dr. N. Asami and Mr. K. Matsui for providing him with this research opportunity. This experiment was conducted in a special Dewar-type colorimetric cell silvered at the top that was developed by Drs. Martin Fleischmann and Stanley Pons. The detailed analysis of the experimental data presented in this report was conducted by Dr. Martin Fleischmann. An independent method of data analysis developed by Dr. Melvin H. Miles while he was in Japan was presented in his NEDO Final Report and shows similar trends for the excess heat effect.

Martin Fleischmann

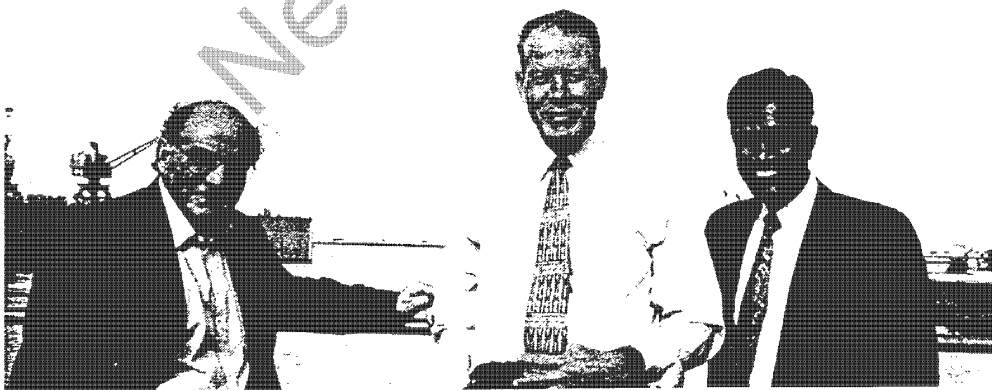
Dr. Martin Fleischmann

Melvin H. Miles

Dr. Melvin H. Miles

Dr. Ashraf Imam

Dr. Ashraf Imam



DRS. FLEISCHMANN, MILES AND IMAM

Executive Summary

The cathode for the heavy water electrolysis experiment was synthesized and processed using elemental palladium and boron (see report # NRL/MR/ 6170-7803) at the Naval Research Laboratory, Washington D.C., USA. The electrolysis experiment, as a part of continuation of the work at Naval Air Warfare Center Weapons Division (see report # NAWCWPNS TP 8302), was carried out by Dr. Melvin H. Miles during his stay at the New Hydrogen Energy (NHE) Laboratory, Sapporo, Japan. In this experiment an ICARUS-2 electrochemical unit and data acquisition system were used similar to those utilized in the earlier work in the IMRA-Europe Laboratories.

The experimental data and the preliminary evaluation [carried out by members of the NHE group] is contained in a series of $(k_R')_{11}$ spreadsheets where $(k_R')_{11}$ is the pseudo radiative lower bound heat transfer coefficient based on the assumption of a zero rate of excess enthalpy generation. The NHE team also quoted a single value of the true heat transfer coefficient determined during the third measurement cycle [Day 3 of the experiment]. The methodology used by the NHE group was non-standard [in the sense that the method used was not that specified for the ICARUS-system]: it appears that this coefficient is related to the "true integral heat transfer coefficient, $(k_R')_{362}$ based on an extrapolation procedure and using the "forward integration" of the experimental data.

It is shown that the value of $(k_R')_{362}$ determined is incorrect: it is evidently too small because it predicts negative rates of excess enthalpy generation even for the third measurement cycle [where $(k_R')_{362}$ was determined] as well as for other measurement cycles in the experimental sequence. A major point of difficulty was evidently the very early development of "positive feedback" [an increase of the rate of excess enthalpy generation with temperature] for this Pd-B cathode. In the general case, the development of such "positive feedback" makes it impossible to evaluate the "true heat transfer coefficients". However, in this experiment the effects were relatively small, and it is shown that such effects can be taken into account in the determination of the "true heat transfer coefficient, $(k_R')_{262}$ " based on the backward integration of the experimental data. The evaluation of $(k_R')_{262}$ was the procedure recommended for use with the ICARUS-systems. A number of further errors in the evaluation given by the NHE group have also been corrected.

It is also shown that the value of $(k_R')_{262}$ agrees with the maximum value of $(k_R')_{11}$ determined in the early parts of the experiment [here on Day 2] before the onset of excess enthalpy generation but after the completion of the exothermic charging of the electrode. It also agrees with the values of $(k_R')_{11}$ determined on Day 61 of the measurement sequence during which there was a near zero rate of excess enthalpy generation.

The use of the correct value of the "true heat transfer coefficient, $(k_R')_{262}$ " shows that there were low levels of excess enthalpy generation throughout the experimental sequence [except Day 61]. The cell was evaporated to dryness on Day 68 of the sequence and, during this day, the specific rate of excess enthalpy generation rose to $\sim 27 \text{ W cm}^{-3}$. Measurements on Day 68 are interpreted in detail, and it is shown that the specific rate of

excess enthalpy generation before evaporation to dryness agrees with that immediately following the completion of this step which is an example of "Heat-after-Death". A further example of such "Heat-after-Death" was observed on the final day of the experiment [Day 69] as well as on the lowering of the current density on Day 26 of the sequence.

Comparisons are made with other experiments principally in a set of footnotes. Suggestions made for further evaluations could form addenda to this report.

Section A

A.1 Introduction

This experiment, designated as FP2-97120402-M7c2 by the New Hydrogen Energy (NHE) group was carried out by M.H. Miles during his stay in 1997-98 in the Sapporo Japan Laboratory of the NHE group. The experiment was started at 10:00 a.m. on December 5, 1997 and terminated at 10:00 a.m. on February 12, 1998. This implies that the experiment was terminated on the 70th day following the start of the experiment. The electrode used had the form of a cylindrical rod of dimensions 4.71 mm in diameter and 20.1 mm in length. This yields:

$$\text{Volume of electrode} = 0.350 \text{ cm}^3 \quad (\text{A.1})$$

$$\text{Area of electrode} = 3.15 \text{ cm}^2 \quad (\text{A.2})$$

This electrode was prepared at the Naval Research Laboratory, Washington, D.C. with a composition of 99.5% Pd+0.5% B (wt%).

Palladium- Boron Cathode

Preparations of the palladium alloy with low boron contents were attempted to keep within the miscibility gap. Three compositions of the palladium-boron alloy were prepared at the Naval Research Laboratory using an arc melter with water-cooled copper hearth. Palladium sponge with five-nine purity and high purity boron were used to produce these alloys. The three compositions had nominal boron concentrations of 0.75, 0.50, and 0.25 weight percent boron. These alloys were identified by their nominal compositions. The glow-discharge mass spectroscopy (GDMS) analyses of the three as-prepared alloys showed boron contents of 0.62, 0.38 and 0.18 weight percent. Processing of the electrodes introduced copper [<26 parts per million by weight], tungsten [<2.2 parts per million by weight], and platinum [<47 parts per million by weight]. The lower boron content alloy had the lowest pick up of these elements. The cast alloys consisting of irregular rod shapes were swaged and rolled to sizes depending on the requirements of the experiment. The samples were annealed for two hours at 650°C resulting in an average grain size of $90 \mu\text{m}$.

X-ray diffraction studies were carried out to characterize the three alloys using a Phillips

diffractometer with a generator setting of 50 kV, 30 mA and a copper target. Two distinct phases of the same cubic structure with different lattice parameters were found in all three alloys. The lattice parameter in one phase remained constant with changes in boron content whereas the lattice parameter of the other phase increased with the increase in boron content. As the boron content increased, the fraction of one phase decreased at the expense of the other phase, as expected. The change in the lattice parameters with boron content occurred in the phase where the starting lattice parameters are the same as pure palladium.

Experimental Details

The Pd-0.5 wt% B electrode was cleaned and polished using the standard diamond paste method at NHE. The specimen was observed using an optical microscope, and the surface looked clean and highly polished with some fine circular polishing lines. The electrode was spot welded on the side to a platinum lead wire. Quick setting epoxy was used to cover the spot welded area, the top of the electrode as well as the end of the glass tubing containing the platinum lead wire. The cell was assembled, placed in the water bath, and connected to the ICARUS 2 system. The cell was then filled with 90 cm³ of 0.1 M LiOD by using a large syringe (50 cm³). It was determined that 82 cm³ of the solution filled the cell to the bottom edge of the silvered portion. This mark was frequently used to determine the amount of D₂O required in re-filling the cell. The D₂O used throughout this experiment was from Isotec, Inc. [99.9 atom% D]. The cell dimensions were 25.0 cm in height with the top 8.0 cm silvered. The outside diameter of the cell was 4.2 cm with an inner diameter of about 2.5 cm.

The cell design [the ICARUS-1 Type] is illustrated in Fig. A.1. The cell number was given as 38 and the experiment was carried out in position A.2 [i.e. in position 2 of thermostat tank A] using an ICARUS-2 Type electrochemical polarization, control and data acquisition system. During the same time period, the Pd-B-Ce alloy prepared by Naval Research Laboratory was run in position A.1 of the thermostat tank and Pd-Ce was run in position A.3 in two other Dewar cells. These experiments are discussed in another report [see ICCF-8 Proceedings]. The electrochemical system consisted of a separate Hi-Tek DT2101 potentiostat wired up as a galvanostat for each cell. These potentiostats/galvanostats are capable of delivering currents of ± 1 A at output voltages up to $\sim \pm 100$ V. A separate potentiostat/galvanostat was also used to deliver constant currents to the resistive heater used to calibrate the cell. The system was controlled by a 486 data acquisition computer which also controlled an Hewlett Packard 44705A multiplexer and data acquisition system. This data acquisition system was on an IEEE-GPIB bus so that it would be anticipated that there would not have been any timing errors introduced into the measurements [see Footnotes C.1 and D.1]. The system is fully described in the associated Handbook, Document Version (2.0) (February 1995)⁽¹⁾ [see Footnotes C.2 and D.2].

The three ICARUS-1 type cells were filled with 90.0 cm³ of D₂O+0.1M LiOD and inserted into a large water thermostat whose temperature was independently controlled by a Techne TE-8A stirrer/heater/regulator unit [see Footnote C.3 and Figure A.5]. The

water thermostat was in turn maintained in a room whose temperature was generally 0 to 2°C higher than that of the water thermostat. This study will focus only on the Pd-B experiment positioned in the center of the bath [position A.2]. It should be noted that the LiOD concentration for this cell was increased to 0.2 M on Day 53 of this study.

At the 7th International Conference on Cold Fusion 19th-24th April 1998 we [i.e. M.H. Miles and M. Fleischmann] decided that a report on M.H. Miles' visit to the NHE Laboratories should include an assessment of the Pd-B experiment using as far as is possible the ICARUS Methodology. We decided that this assessment should include:

- (i) comments on differences between the execution of the experiments carried out in the NHE Laboratory and the procedures recommended for use with the ICARUS-1 and ICARUS-2 Systems ^{(1),(2)};
- (ii) a commentary on the calibration of the ICARUS-1 Type cell used in the Pd-B experiment;
- (iii) an assessment of the presence [or absence] of excess enthalpy generation in the Pd-B experiment;
- (iv) an investigation of the presence [or absence] of the effects of "positive feedback"; [e.g. see ^{(6),(7),(8)}];
- (v) An investigation of the presence [or absence] of the effects of "Heat after Death"; [e.g. see ^{(9),(10)}];
- (vi) an assessment of the implications of the evaluations of the Pd-B experiment for further research in this field.

There are also some secondary reasons for choosing the Pd-B experiment for further detailed evaluations [see Footnote C.4].

This Report is divided into the following Sections:

- Section A:** the main text.
- Section B:** a description of the methods of data evaluation as characterized by the relevant heat transfer coefficients.
- Section C:** a set of Footnotes and Comments relating to Sections A and B.
- Section D:** a further set of Footnotes and Comments, which perhaps should have a restricted circulation.
- Section E:** A set of footnotes and comments whose circulation should be restricted.

A.2 Some Important Preliminary Definitions, Descriptions and Evaluations

The various radiative heat transfer coefficients used in this report are defined as

$$(k_R')_{i,j,l}$$

where R is radiative heat transfer, i = 1,2,3 denotes "differential", "backward integration" and "forward integration" respectively, l=1,2 denotes "lower bound" and "true"

respectively [see Footnote C.12] and j , if used, denotes the time period of the measurement cycle as given below

- $j=5$, times somewhat above the origin
- $j=6$, times somewhat above t_1 [application of calibration pulse]
- $j=7$, times somewhat above t_2 [cessation of calibration pulse]
- $j=8$, combination of times for $j=6$ and $j=7$

These values of " j " cover time periods where the cell temperature shows large changes. In addition, the radiative heat transfer coefficient $(k_R'o)_{i,j,l}$ is also frequently used and is obtained from extrapolations of the straight line forms of the calorimetric equations [see Equation B.11 and B.12].

The protocol used for the experiment was as follows:

- (i) the electrode was first of all polarized for two days without any application of calibration pulses;
- (ii) on the third day [and on all subsequent days including days 68 and 69 when the cell had reached dryness] calibration pulses were applied;
- (iii) changes of current density were made frequently at times close to 10:00 am of the measurement cycles; these changes of current density are shown in Table A.1 and in Fig. A.2B found at the back of the text;
- (iv) the cell was "topped up" with D_2O whenever this was judged to be necessary at 10:00 am, the start time of all the measurement cycles; the cell was then left to equilibrate for 9 hours followed by the application of calibration pulses of 6 hour duration [the start time of all the calibration pulses was 7:00 pm]; the cell was then again left to equilibrate for a further 9 hours before reaching 10:00 am of the next day of the experimental sequence;
- (v) as is evident from (iv), the duration of the measurement cycles was 24 hours; the addition of D_2O and the changes in the current density were always made by M. H. Miles at his discretion. Everything else was controlled by the NHE computer program.

This protocol differs substantially from that specified for the operation of the ICARUS-1 and ICARUS -2 Systems, which was as follows ^{(1),(2)};

- (ia) the electrodes were to be polarized for 4 days [i.e. two measurement cycles, see (va) below] without any application of calibration pulses;
- (iia) on the 5th day [i.e. for the third measurement cycle] and for nine further measurement cycles calibration pulses were to be applied as specified in (iva) below; this was to be followed by two further measurement cycles without the application of calibration pulses and, in turn, by ten further cycles with calibration pulses. A total experiment duration of 48 days was therefore specified for the initial phase of the work.

- (iiia) the initial experiments were to be carried out at a single, low current density, typically $< 250 \text{ mA cm}^{-2}$ or 788 mA for this Pd-B electrode; in later experiments a single, low current density was to be applied for various initial durations followed by a raising of the current density to values typically $> 1 \text{ A cm}^{-2}$; this protocol was in broad accord with that used in previous investigations ^{(15),(16)}; changes of current density were to be made at the beginning of the measurement cycles. The large size of the Pd-B electrode [$A=3.5 \text{ cm}^2$] limited the current density to about 320 mA/cm^2 .
- (iva) cells were to be "topped up" at the start of each measurement cycle; the cells were then to be left to equilibrate for 12 hours and calibration pulses of 12 hour duration were then to be applied; the cells were then again to be left to equilibrate for a further 24 hours so as to reach the start of the next measurement cycle.
- (va) as is evident from (iva), the duration of the measurement cycles was to be 48 hours.

Further important aspects of the protocols specified for the ICARUS-1 System are given in Footnote C.5. These protocols were designed with two major aims: firstly, to verify the modeling of the cells which is the basis of the methods of data evaluation; secondly, to facilitate these methods of data evaluation. Both of these aspects are outlined in Sections B.2 and B.3 together with the associated Comments and Footnotes.

Needless to say, with the progression of the work, parts of these protocols were expected to become redundant. Furthermore, the protocol (ia) - (va) had been designed for the execution and evaluation of appropriate "blank experiments," and it was unlikely that this protocol would be entirely suitable for experiments on the Pd-D₂O type systems [especially experiments in which there were changes in the current density]. We also have to take special note of an unsatisfactory situation. With the exception of some fragmentary analyses of data sets collected during the start-up of the NHE project ⁽³⁾ we have, as of now, no comprehensive data sets and evaluations of "blank experiments" [see Footnote C.27]. We therefore have to rely on experiments carried out at IMRA Europe and on the analyses of simulations e.g. see Reference 13 for this aspect of the work; i.e. we are unable to investigate/validate the operation of the ICARUS-1 and -2 Systems installed in Sapporo.

As far as the second aspect is concerned, we have to take note of the fact that in view of the changes in the protocols made by both M. H. Miles and the group at NHE [(iv) and (v) as against the specified (iva) and (va)], we are unable to apply the simplest method of calibration leading to $(k_R')_2$ [see Section B.2] in view of the inadequate relaxation of the temperature following the application of the heater pulses [see Footnotes C.28 and C.29].

We now consider further the major differences between the operation of the Pd-B

experiment and the conditions used in previously reported investigations, e.g. see References 12, 15 and 16. Apart from the frequent changes of current density [see Footnote C.30, Fig. A.2B], we can see that these current densities were mostly in the vicinity of the threshold value required for the onset of the phenomenon of excess enthalpy generation ⁽¹²⁾ [see Footnote C.31]. Furthermore, the cell temperatures were mostly below the level required for the onset of "positive feedback", Fig. 2.A ^{(6),(7),(24)} and which leads to a marked increase in the rates of excess enthalpy generation [see Footnotes C.32 and C.33]. The conditions in the cell therefore remained in the vicinity of the region of onset of "positive feedback" and, under these conditions we would not expect to see a marked build up in the rate of excess enthalpy generation [see Footnote C.34].

Consideration of Fig. A.2B also allows us to decide on the measurement cycles likely to provide examples of "Heat-after-Death" [objective (v) of this investigation] as was pointed out in the original investigation ^{(9),(10)}. It would be expected that this phenomenon would be observable under several distinct conditions, which include [using the original classification numbers]:

- Cell full: cell operated at intermediate temperatures; cell current then reduced in stages; [original number 1]
- Cell empty: cell allowed to boil dry; cell then maintained at the rail voltage of the galvanostat; [original number 5]
- Cell empty: cell allowed to boil dry; cell disconnected from the galvanostat. [original number 6]

Consideration of the "hard copy" of the data sets shows that condition #5 applies to part of Day 68 of the sequence measurement cycles [see Section A.8] while condition #6 applies to part of Day 69 of this sequence [see Section A.9]. Consideration of Fig. A.2B shows that condition #1 is likely to apply to several of the measurement cycles. The effects would be expected to be most marked for Day 26 [reduction of the cell current from above to below the threshold for excess enthalpy generation; reduction in cell temperature from above the level for the onset of "positive feedback" to below this level]. Attention is confined in this Report to this particular day [see Section A.10] although it is evident that there are several further regions of time which might well give examples of "Heat-after-Death" following Scenario 1.

In this Section we should also consider a further difference between the protocols for Pd-B experiment and those used in earlier studies; namely, the schedules of addition of D₂O to make up the losses due to electrolysis. Table A.1 and Fig. A.3 illustrate the effect on the change in volume of the electrolyte in a cell in an hypothetical experiment carried out first at a cell current of 200 mA for 29 days followed by a cell current of 500 mA and with a daily schedule of D₂O additions. It can be seen that the mean volume of the electrolyte falls by some 1.21 cm³ between the two time regions. We can estimate that this would cause a decrease of the mean value of $(k_R')_{12}$ by ~ 0.15% or of $(k_R')_{22}$ by ~

0.075%. Such small changes are close to the error limits quoted for the instrumentation and can normally be neglected. However, the magnitude of the changes are above the error limits which can actually be achieved [e.g. see ⁽¹³⁾ and Section A.4] and should be taken into account in evaluations carried out at the maximum achievable precision and accuracy.

Fig. A.4 shows the effects of D₂O additions as actually used in this Pd-B experiment. This figure assumes that the D₂O is lost only by electrolysis. Experimental measurements, however, show an additional 4% of the D₂O was carried out of the cell by the gas stream. The exact volumes of D₂O were recorded throughout this experiment [see Footnote C.35]. It can be seen that the expected changes in $(k_R')_{12}$ due to the electrolyte volume now lie between -0.3 and +0.6% and of $(k_R')_{22}$ between -0.15 and +0.3%, changes which should certainly again be taken into account. These numbers will change somewhat due to the additional loss of D₂O by the gas stream [see Footnote C.36].

The schedule of additions leads to an important conclusion. We find that by Day 67 the total volume of D₂O added was 262.5 cm³ whereas the total volume electrolyzed was 253.3 cm³. Earlier in this experiment on Day 39, the total volume of D₂O added was 112.5 cm³ whereas the total volume electrolyzed was 108.2 cm³. It is evident that the volume of D₂O added is consistently about 4% larger than the volume electrolyzed and, therefore, there could not have been any recombination of the deuterium and oxygen produced by electrolysis. This is in agreement with earlier measurements ⁽¹²⁾ [see Footnote C.37] and numerous measurements by other authors [see Footnote C.38].

The horizontal lines in Figs. A.3 and A.4, delineate the volumes of D₂O below and above which we would expect the electrolyte level to fall below the base of the silvering in the upper part of the cell, Fig. A.1, or to approach the base of the Kel F plug at the top of the cell. The results of Figure A.4 are calculated theoretically by assuming that D₂O is removed only by electrolysis. Experimentally, the electrolyte was observed to fall below the base of the silver line prior to the D₂O addition only on Day 25 (-1 cm³), Day 41 (-1 cm³), Day 48 (-2 cm³), Day 50 (-1.5 cm³), Day 53 (-0.5 cm³), Day 55 (-2 cm³), and Day 67 (-6 cm³). It can be seen that the electrolyte level remained within the space defined by this silvered portion throughout most of the measurement cycles [see Footnote C.39]. However, we can see that the electrolyte level may have approached the base of the Kel F plug at the start of several of the measurement cycles following the "topping up" of the cells. In work carried out at IMRA Europe, it has been established that such "overfilling" of the cells leads to an anomalous increase of the pseudo radiative heat transfer coefficients by 4 - 5% of the values which apply at the mean. This increase in (k_R') is almost certainly due to an increase in the conductive contribution through the Kel F plug to the overall heat transfer from the cell [c.f. equation (B.2)]. Experimentally the "overfilling" of the cell in this Pd-B experiment occurred on Day 26 (92.0 cm³), Day 43 (94.0 cm³), Day 50 (93.5 cm³), and on Day 57 (94.0 cm³). This was done intentionally because of long weekends when it was not possible to enter the laboratory.

This type of "overfilling" behavior may apply also to Day 61 which is a measurement cycle for which we can get important confirmatory evidence of the "true heat transfer

coefficient" which applies to the operation of the cell [see Fig. A.20, Section A.5]. Fig. A.20 shows the expected increase in $(k_R')_{11}$ at times close to the "topping up" of the cell. Another explanation is that the large addition of D_2O (9.0 cm^3) at room temperature [23°C] on Day 61 suddenly cooled the cell contents, thus $(k_R')_{11}$ is too large during the cell's recovery from this cooling shock.

Finally, we also make a number of preliminary assessments of the form of the temperature-time and cell potential-time series for Day 3, i.e. the third measurement cycle of this experiment, Figs. A.5 - A.7. The data for this day are of special importance because the group at NHE has quoted a value of the "true heat transfer coefficient" as given by their method of evaluation for this day. This value of the "true heat transfer coefficient" was then used in the evaluation of all the measurement cycles. The evaluation by NHE is considered further in Section A.3 while Section A.4 gives the application of the ICARUS Methodology to this particular data set.

Columns 1 - 6 of Table A.2 give the "raw data" for the third measurement cycle [the remaining columns are discussed in Section A.3]. We can immediately draw a number of important conclusions. Thus Fig. A.5 gives a plot of the temperature of the water bath versus time for the first 32,400 s of the measurement cycle [the period $0 < t < t_1$ preceding the application of the heater calibration pulse] while Fig. A.6 gives plots of the cell temperature versus time for the same period and for both positions in the cell where the temperature was measured, see Fig. A.1. It is evident that the "noise" level of the measurements in the water bath [$\sigma = 0.0088 \text{ K}$, mean = 295.198 K] is much higher than that of the measurements of the cell temperature, Fig. A.6. This difference [$\sigma = 0.0027 \text{ K}$] is to be expected because the water bath is controlled by a single thermal impedance whereas the cell is controlled by two impedances in series. At the same time, the "noise" in the temperature of the water bath is much higher than that in the original measurements with the ICARUS-Systems [$\sigma = 0.003 \text{ K}$] ⁽¹²⁾ and, in our experience, such an increase is due to inadequate control of the room temperature. The room temperature was usually 0-2 K above the bath temperature, i.e. 22 to 24°C . Typical variations in the temperature of this room are shown elsewhere [Journal of Electroanal. Chem., Vol. 482, pp. 55-65, 2000].

It will be evident that the "noise" in the measurements of the temperature of the water bath is one factor which will limit the precision of the "lower bound heat transfer coefficients, $(k_R')_{11}$ via its effect on the temperature function, $f_1(\theta)$, see Section B.3. The value $\sigma = 0.0088 \text{ K}$ is outside the range specified for the ICARUS-1 system if measurements are made at low cell temperatures, Table A.2. By contrast, the "true heat transfer coefficients" are not affected by such fluctuations because the temperature function $f_2(\theta)$ [see Section B.3] is determined by the cell temperature alone.

It can be seen that the variation with time of the cell temperature measured at the two positions in the cell, is systematic, Fig. A.6. Moreover, it is clear that there is a systematic difference in temperature between the two positions, which must be due to either one or two errors in the calibration [see Footnote C.40]. For these measurements

we obtain mean $[\theta_{\text{short thermistor}} - \theta_{\text{long thermistor}}] = 0.0045 \text{ K}$ and $\sigma[\theta_{\text{short thermistor}} - \theta_{\text{long thermistor}}] = 0.0027 \text{ K}$. The mean gives an indication of the accuracy in $(k_R')_{11}$ which we can expect to achieve. The error $\sim 0.05\%$ is somewhat above the target for the precision of the measurements, errors $< 0.01\%$, which is hardly surprising. The standard deviation gives double the value of the expected standard deviation for the measurements with one thermistor. We can see that this value, $\sim 0.00135 \text{ K}$, will not affect the accuracy of the determination of any version of the "true heat transfer coefficient". However, we should note that it is evidently desirable to calibrate the thermistors so that we can make the temperature measurements to within $\pm 0.001 \text{ K}$ [see Footnote C.41].

Differences in temperature between those given by the "short thermistor" and "long thermistor" will be considered further in Section A.7 dealing with Day 68 as the cell is being driven to dryness and in Section A.9 dealing with "Heat-after-Death" on Day 69.

Finally, we consider the plots of the "raw data", for Day 3, Fig. A. 7. We can see immediately the inadequacy of restricting the calibration pulse to 6 hours because the temperature has not relaxed to equilibrium in this time period [see Footnote C.42]. However, in this particular case there is an evident complication because of the very early establishment of "positive feedback". This effect can be seen most directly from the delayed relaxation of the temperature to the base line following the cessation of the heater calibration pulse [the base line is given by the extrapolation of the $\theta - t$ series observed before the application of the calibration pulse]. Evidently, the raising of the cell temperature by the calibration pulse has led to an increase in the thermal output from the cell which persists following the termination of the calibration, i.e. a form of "positive feedback" [see Footnote C.43]. The calibration of such a system can obviously only be achieved with many restrictions and with great difficulty [see Footnote C.44].

The interpretation of Fig. A.7 will be considered further in Sections A.3 and A.4.

A.3 The NHE Interpretation of This Pd-B Experiment (M7c2)

As has already been pointed out, the NHE interpretation of the Pd-B experiment rests on the determination of the "true heat transfer coefficient" on Day 3 of the measurement cycles. Apart from the citation of the value of this coefficient ($0.793504 \times 10^{-9} \text{ WK}^{-4}$) in the header for the spreadsheet for Day 1, the information given by NHE is contained in a set of spreadsheets, which appear to be related to the $(k_R')_{11}$ spreadsheets of the ICARUS Methodology for analyzing the data. The description of these spreadsheets is contained in Footnote C.45. In addition to the description contained in this Footnote, we have to take note of the following observations:

- a) it is not clear how the value of the "true heat transfer coefficient" was determined nor which of the definitions of the heat transfer coefficients may have been used [see Section B.3]. The $(k_R')_{21}$ - spreadsheets [or $(k_R')_{31}$ spreadsheets] ^{(1),(2)} which would have allowed an investigation of this value [and which were part of the ICARUS data processing system] were not given [see also Section A.4]. However, it is likely that this was

the coefficient $(k_R')_{32}$ and it will be assumed here that this was the case, i.e. we will assume that the values of the excess enthalpies given in columns (12) and (18) of the spreadsheets were based on calculations using the single value

$$(k_R')_{32} = 0.793504 \times 10^{-9} \text{ WK}^{-4}$$

- b) it is also not clear to what extent the values of the "true heat transfer coefficient" and of the excess enthalpies may have been affected by the value

$$C_p M = 490 \text{ JK}^{-1}$$

used in the calculations. Values as high as this applied to cells used prior to 1992, and the Handbooks for the ICARUS Systems contained instructions for changing this [and other] parameter(s) depending on the value found using the methods of evaluation outlined in the Handbooks ^{(1),(2)} [see also Sections A.4 and B.3].

It should be noted that the "guesstimate" of the water equivalent of the cell is:

$$C_p M \sim \text{contribution of D}_2\text{O in the electrolyte} + \text{contribution of the glass in the inner cell wall} = (419 + 31) \text{ JK}^{-1} = 450 \text{ JK}^{-1}$$

The remaining components of the cell [LiOD, metals, glass framing, heater, thermistor, a proportion of the Kel-F plug] will contribute only a small additional term to $C_p M$. It follows, therefore, that observations of $C_p M$ far above or below 450 JK^{-1} indicate malfunctions of the methods of data evaluation.

- (c) as has been noted elsewhere [see Section B.2], the values of the rates of evaporative cooling, columns (9) and (16), given in the spreadsheets cannot be calculated using the instructions given in the Handbooks for the ICARUS-1 and -2 Systems ^{(1),(2)}. The differences are not important at low temperatures [such as those which apply to Day 3 of the measurement cycles, Table A.2] but become significant at temperatures close to the boiling point. However, at such elevated temperatures other factors neglected in the calculations carried out by NHE become even more important [see Sections B.2 and A.7].
- (d) it is apparent that the enthalpy inputs given in column 8 of the NHE spreadsheets have been calculated using 1.54 V as the thermoneutral potential, E^0_H , whereas most other authors have used the value 1.527 V. The circumstances leading to our choice of the value 1.54 V have been described elsewhere [see Footnote C.46]. The value 1.527 V has been

used in deriving Tables A.3 - A.9).

While considering the values of the enthalpy inputs, column (8) of the NHE spreadsheets, it should be noted that these have been given to 4 significant figures whereas they should have been given to 5 significant figures. It is not clear at the present time whether the values of $(k_R')_{11}$ listed in the spreadsheets were calculated using the enthalpy inputs rounded to 4 figures or whether a higher precision was maintained in the calculations.

- (e) the most serious shortcoming of the NHE calculations is that the input due to the calibration heater has been entered as zero rather than the actual value given separately as 0.25000 W [see Footnote C.47]. In the procedure used by NHE ⁽¹¹⁾ the "lower bound heat transfer coefficient, $(k_R')_{11}$ " is calculated with this assumed zero enthalpy input, and it is then assumed that the magnitude of the enthalpy input, can be recovered together with any rate of excess enthalpy generation by using this derived "lower bound heat transfer coefficient" together with the "true heat transfer coefficient, $(k_R')_{32}$ and $f_1(\theta)$. Let us assume first of all that such a procedure is correct. Then we can see an immediate disadvantage as compared to the method outlined for the ICARUS Systems in that we are unable to determine whether $(k_R')_{11}$ during the period of the application of the calibration pulse in $t_1 < t < t_2$ is the same as for $t < t_1$, or $t > t_2$ [see Footnote C.48]. The data derived, e.g. see Fig. A.8 below, are certainly further degraded by using an incorrect value of $C_p M$. However, in actual fact, the procedure used by NHE is invalid as has been pointed out in a Report ⁽⁴⁾ and in subsequent correspondence [see Footnote C.49]. It is difficult to see why the straightforward procedure outlined in the Handbook for the ICARUS-1 System ⁽²⁾ was not followed.

Notwithstanding the reservations (a)-(e) about the evaluations carried out by NHE, columns 7-10 of Table A.2 summarize these evaluations [columns 12, 14, 18 and 19 of the $(k_R')_{11}$ spreadsheets given by NHE] while the derived data in columns 11 - 14 of Table A.2 are also based on the NHE evaluations. Columns 15 - 17 of Table A.2 make a correction for (e); the remaining required corrections have been made for all the derived data given in Tables A.3-A.9, Sections A.4-A.7 and A.10.

We consider next the values of the "lower bound heat transfer coefficient, $(k_R')_{11}$," compared to the "true heat transfer coefficient" as given by the NHE evaluation, Columns 7 and 9 of Table A.2 and Fig. A.8, as well as the values of the rates of excess enthalpy generation, columns 8 and 10 and Fig. A.9. Table A.2 also includes the 11-point averages of $10^9 (k_R')_{11}$ and Q_{excess} in columns 13 and 11 and the further 6-point averages of $10^9 (\overline{k_R'})_{11}$ and $\overline{Q_{\text{excess}}}$ in columns 14 and 12 respectively using the data evaluated with the $\theta - t$ series as given by the "short thermistor". We can

only conclude from these data that the evaluations are incorrect based on the following evidence:

- (f) it is impossible for the "true heat transfer coefficient, $(k_R')_{32}$ " to be smaller than the "lower bound heat transfer coefficient, $(k_R')_{11}$ " because the lower bound value is based on the assumption that there is a zero rate of excess enthalpy generation in the cell. The type of difference seen in Table A.2 and Fig. A.8 could only arise if the cell was endothermic, and the endothermicity has already been fully taken into account using the thermoneutral potential. Any additional endothermicity, therefore, requires that the cell operates as a spontaneous refrigerator, and this violates the Second Law of Thermodynamics.
- (g) the pronounced variation of the "lower bound heat transfer coefficient, $(k_R')_{11}$ " with time following the application of the heater calibration pulse at $t = t_1$ and its cessation at $t = t_2$ implies at the very least that the "raw data" have been evaluated using an incorrect value of the water equivalent, $C_p M$, of the cell.
- (h) the excess enthalpy given by the NHE evaluation is apparently negative both for $t < t_1$ and $t > t_2$ which is a further illustration of the apparent violation of the Second Law of Thermodynamics, cf. (f) above.
- (i) it has been maintained ⁽¹¹⁾ that the NHE evaluation recovers the magnitude of the heater calibration pulse, ΔQ , during its period of application, $t_1 < t < t_2$, together with any rate of excess enthalpy generation. Fig. A.9 shows that this is incorrect: the values of the rates of excess enthalpy generation [which here include the enthalpy input to the calibration heater] are less than ΔQ in the period $t_1 < t < t_2$ if we take $Q_{\text{excess}} = 0$ as the base line. If we fix the base-line at the level of the negative rate of excess enthalpy generation for $t < t_1$, then $Q_{\text{excess}} > \Delta Q$ during the period of the calibration pulse, $t_1 < t < t_2$ [see Footnote C.50].

We conclude that the evaluation given by NHE is invalid and that it is likely that this evaluation is subject to several distinct errors.

A.4 ICARUS Type Interpretation of the Experiment

As a first step, we correct the $(k_R')_{11}$ spreadsheet, Table A.2, by including the magnitude of the calibration pulse, ΔQ , in the definition of the "lower bound heat transfer coefficient". The modified values of $10^9 (k_R')_{11}$ are shown in column 15 while the values of $10^9 (\overline{k_R'})_{11}$ and $10^9 (\overline{\overline{k_R'}})_{11}$ are given in columns 16 and 17. The values of $10^9 (k_R')_{11}$ in the region $t_1 < t < t_2$ can now be shown together with those for $t < t_1$ and $t > t_2$ on a graph using a single scale for the ordinate, Fig. A.10. While we cannot be certain whether or not an incorrect choice of $C_p M$ can explain the fall of $(k_R')_{11}$ in the region $t > t_1$

[but close to this time] or the rise for $t > t_2$ [but close to this time], it is clear that $(k_R')_{11}$ drops markedly in the region $t_1 < t < t_2$ compared to the values for $t < t_1$ and $t > t_2$. Such a drop in $(k_R')_{11}$ can only be due to the neglect of the build up of the rate of excess enthalpy generation during $t_1 < t < t_2$. It follows that the increase in temperature due to the calibration pulse increases the rate of excess enthalpy generation. In fact, the experiment shows a very early establishment of "positive feedback" as is indeed evident from the plot of the "raw data", Fig. A.5 [see Footnote C.51]. It is very important that the presence of "positive feedback" can be established by a simple examination of a $(k_R')_{11}$ spreadsheet constructed according to the instructions in the ICARUS-1 Handbook ⁽²⁾.

It should be noted that the amplitude of the calibration pulse would have had to be $\Delta Q = 0.2763$ W in order to bring the values of $(k_R')_{11}$ in the region $t_1 < t < t_2$ to the level of the regression line which applies to the data for $t < t_1$ and $t > t_2$. Such a change in ΔQ is beyond all possibilities.

The next step is to prepare a modified $(k_R')_{11}$ spreadsheet where we correct the enthalpy inputs [see (c), (d) and (e) in Section A.3] and present the data in a form suitable for the application of equation (B.12) columns 3 and 4 of Table A.3, while column 5 gives the result of the application of equation (B.12). In view of the early intervention of "positive feedback", we would only expect to be able to apply equation (B.12) at times close to t_1 where we see that the "true heat transfer coefficient", $(k_R')_{11}$ must be at least $0.83808 \times 10^{-9} \text{ WK}^{-4}$ while the water equivalent, $C_p M$ is of the order of 454 JK^{-1} [in agreement with the "guesstimate", see Section A.3].

The influence of "positive feedback" on the failure of simple methods for the evaluation of the "lower bound" and "true heat transfer coefficients" as well as of the water equivalent of the cell is also shown clearly by attempts to derive $(k_R')_{181}$ [which rely on the combination of data for the time regions $t_1 < t < t_2$ and $t_2 < T$, (see Section B)]. This evaluation has been found to be especially useful in the analyses of data sets for "blank" experiments [e.g. see ⁽¹³⁾]. Fig. A.11 illustrates that we are unable to obtain a satisfactory interpretation of such data for the experiment as is also shown by column 6 of Table A.3.

Column 7 of Table A.3 gives the values of $10^9 (k_R')_{11}$ while column 8 gives the running mean, $10^9 (\overline{k_R'})_{11}$, of column 7. We can see that these values do not differ greatly from the corresponding values in columns 9 and 15 of Table A.2 which, are based on the correction of (e) alone [see Section A.3] of the NHE analysis. Fig. A.12 gives the plot of the data in column 7 versus time and also shows the variation of $10^9 (k_R')_{11}$ with time predicted using the values for $t < t_1$ and the known behavior established with appropriate "blank" experiments e.g. ^{(4),(13),(14)}. As in the case of the data in Fig. A.10, we can see that the temperature rise induced by the calibration pulse leads to a decrease in $(k_R')_{11}$ while the cooling consequent on the termination of the pulse leads to an increase in $(k_R')_{11}$. These changes can only be due respectively to an increase and decrease in the rate of excess enthalpy generation, which cannot be taken into account in deriving the values of $(k_R')_{11}$, i.e. the effects of "positive feedback".

Fig. A.12 shows that we still observe discontinuities in the "lower bound heat transfer

coefficient, $(k_R')_{11}$ at t_1 and t_2 . However, it is evident that there can be no mechanism, which could account for such changes, which must therefore be due to an error in the analysis. The most obvious error is the use of an incorrect value of $C_p M$ [see (b), Section A.3]. The analysis of the time dependence according to equation (B.12) in the region $t > t_1$ [but adjacent to t_1] indicates that the correct value is $\sim 450 \text{ JK}^{-1}$, and columns 9, 10 and 11 of Table A.3 give respectively $C_p M (d\Delta\theta/dt)$, $10^9 (k_R')_{11}$ and $10^9 (\bar{k}_R')_{11}$ based on this value of $C_p M$. Fig. A.13 shows a plot of the values in column 10 versus time, and we can see that the discontinuities in the heat transfer coefficient at $t > t_1$ and $t > t_2$ [but adjacent to these times] are now eliminated. However, as expected, the effects due to "positive feedback" are maintained.

Column 12 of Table A.3 also gives values of the rates of excess enthalpy generation calculated with the "true heat transfer coefficient, $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ established using the $(k_R')_{21}$ spreadsheets and other confirmatory evidence outlined in Section A.5 below. Fig. A.14 shows that there is indeed only a small rate of excess enthalpy generation for $t < t_1$ while the application of the calibration pulse leads to a build-up of this rate which again decreases for $t > t_2$ [there is a small long-term increase in the rate of excess enthalpy generation for $t > t_2$]. Fig. A.15 shows a similar calculation but using the NHE methodology [note the difference in scales of the y-axes in Figs. A.14 and A.15]. We again see a near zero rate of excess enthalpy generation for $t < t_1$, while for $t > t_1$ but adjacent to t_1 we now see the step due to the calibration pulse, $\Delta Q = 0.2500 \text{ W}$. In the region $t_1 < t < t_2$, we then see the build-up in the rate of excess enthalpy generation due to "positive feedback". At $t = t_2$ but adjacent to t_2 we again see a step in the total observed rate of excess enthalpy generation. As expected, this step again corresponds to the expected value $\Delta Q = 0.2500 \text{ W}$; at longer times we see the gradual decrease of the rate of excess enthalpy generation due to the removal of the effects of "positive feedback".

The comparison of the interpretation of the $(k_R')_{11}$ spreadsheet prepared according to the instructions for the ICARUS Systems ^{(1),(2)} with that prepared using the NHE methodology, Section A.3, illustrates the importance of following the instructions laid down in the Handbooks ^{(1),(2)}. We can also see that a great deal of information about the behavior of the systems can be derived from the interpretation of the correctly evaluated $(k_R')_{11}$ spreadsheets and these spreadsheets serve as the basis for the next stage of the analysis of the measurement cycles.

This next, third, step in the data analysis is the preparation of the $(k_R')_{21}$ spreadsheet for the whole measurement cycle shown in Table A.4 which can be used equivalently to produce the $(k_R')_{31}$ spreadsheet [Table A.4 is an abstract of the two spreadsheets] [see Footnote C.52]. In evaluating the integrals used in the preparation of this spreadsheet, it has been noted that t_1 and t_2 correspond closely to the relevant measurement intervals [this is not illustrated in this Report]. The integrals have been evaluated using the trapezium rule following the insertion of additional data points t_1 and t_2 [see Footnote C.53].

It can be seen that Table A.4 and a comparison of the plots of $(k_R')_{21}$ and $(k_R')_{31}$ versus time, Fig. A.16, with the corresponding plots for "blank experiments", e.g. see ^{(4),(5),(14)},

shows very clearly the intervention of "positive feedback" due to the superposition of the calibration pulse. If we focus attention first of all on the behavior of $(k_R')_{31}$ for $t < t_1$, then we see the expected small decrease with increasing time [see Footnote C.54]. For $t > t_1$ we see a more rapid decrease due to the onset of "positive feedback". The effects of this "positive feedback" decrease for $t > t_2$, so that we observe a small increase of $(k_R')_{31}$ with increasing time in this region.

The variation of $(k_R')_{21}$ with time can be interpreted in a similar way provided one bears in mind that there is now no region in time in which the integrals used in the calculation of the heat transfer coefficient are independent of the effects of "positive feedback". This influence of "positive feedback" on the integrals used in the evaluation of $(k_R')_{21}$ explains why we cannot obtain a satisfactory evaluation of the target value of the "lower bound heat transfer coefficient, $(k_R')_{261}$ " column 5 of Table A.4. We would only expect to be able to apply the ICARUS methodology in a region of time where the influence of "positive feedback" can be expected to be adequately small, say in the region 72,300 - 75,300 s of the measurement cycle. The estimates of the "lower bound heat transfer coefficient, $(k_R')_{261}$ " and of $C_p M$ are shown in bold type in column 5 of Table A.4.

Again, if we bear in mind the influence of "positive feedback" on the integrals, we would only expect to be able to apply the ICARUS methodology to the initial part of the calibration period to give estimates of the target value $(k_R')_{361}$ [say between 32,400 and 35,400 s in Table A.4]. These estimates of the "lower bound heat transfer coefficient, $(k_R')_{361}$ " and of $C_p M$ are shown in bold type in Column 10 of Table A.4.

It is important here to draw attention to a restriction, which we have applied to the evaluation of the integral heat transfer coefficients. Table A.4 shows that abscissa in columns 2 or 7 are always numerically small compared to the ordinates in columns 3 or 8. In consequence the heat transfer coefficients $(k_R')_{21}$ and $(k_R')_{31}$ in columns 4 or 9 can be evaluated with good precision notwithstanding the limited accuracy of the values of the water equivalents, $C_p M$.

This effect becomes much more important when we come to consider the evaluation of the "true heat transfer coefficients, $(k_R')_{262}$ and $(k_R')_{362}$ [see the discussion of Table A.5 further below]. For the particular case of the experiment we see that we must rely on the evaluation of the "true heat transfer coefficient, $(k_R')_{362}$ in view of the intervention of "positive feedback" even though this heat transfer coefficient is not the target value of the ICARUS methodology [see Footnote C.55]. Columns 11 and 12 of Table A.4 now show that the abscissae and ordinates have comparable magnitudes in the time range in which we might conceivably be able to neglect the effects of "positive feedback" and the evaluation of $(k_R')_{362}$ therefore fails.

The comments which have been made about the evaluation of the integral heat transfer coefficients using the whole measurement cycles apply equally to the evaluations according to the instructions and software in the ICARUS Systems ^{(1),(2)}, Table A.5. The evaluation of $(k_R')_{261}$ [not shown in Table A.5] fails because of the intervention of "positive feedback" and the precision of $(k_R')_{31}$ and $(k_R')_{361}$, columns 4 and 5 of Table A.5

is low because of the need to restrict attention to the region $t > t_1$ but close to t_1 . This is equally true of the accuracy of $(k_R')_{32}$ and $(k_R')_{362}$ shown in columns 8 and 9 of Table A.5. However, the evaluations of these coefficients are instructive because it is virtually certain that the value of the "true heat transfer coefficient" quoted by NHE is either the value of $(k_R')_{32}$ at a particular time or else $(k_R')_{362}$ evaluated over a particular range of time.

Columns 10 - 13 of Table A.5 illustrate the evaluations of $(k_R')_{22}$ and $(k_R')_{262}$ and it is clear that we cannot apply the ICARUS Methodology as set out in the Handbooks ^{(1),(2)} in view of the intervention of "positive feedback". We therefore have to investigate whether we can modify the approach so as to allow the determination of the "true heat transfer coefficients". We have to note that it is unlikely that we would be able to find a generally valid procedure because it is in general not possible to calibrate closed loop systems subject to "positive feedback". However, for the particular example of Day 3 of the experiment we can see that the effects of "positive feedback" are relatively small and, moreover, confined in the time-domain, Fig.A.14. We can therefore include the observed values of the rates of excess enthalpy generation in the evaluation of the integral of the enthalpy input and use this modified integral to re-evaluate $(k_R')_{22}$ and $(k_R')_{262}$ columns 14 - 16 of Table A.5. Fig. A.17 illustrates this evaluation. It can be seen that we do indeed now obtain a satisfactory fit to equation (B.24), which explains the choice of

$$(k_R')_{262} = 0.85065 \times 10^{-9} \text{ WK}^{-4} \quad \text{and} \quad C_p M = 450 \text{ JK}^{-1}$$

for the further evaluation of the data.

In view of the fact that this evaluation of the "true heat transfer coefficient, $(k_R')_{262}$ requires the development of a special approach, it is necessary [and advisable] to investigate whether the value obtained can be confirmed by other means using different parts of the experiment [i.e. other measurement cycles]. Such confirmations can be obtained using the measurements on Day 61 and the first 57 hours of Days 1 and 2. These confirmations are outlined in Sections A.5 and A.6 respectively.

A.5 Application of the ICARUS Type Interpretation to the Data for Day 61

It has been shown in Sections A.3 and A.4 that the early intervention of "positive feedback" requires us to modify the ICARUS evaluation strategies in order to achieve the calibration of the system i.e. to determine the value of the "true heat transfer coefficient". It is therefore important to find confirmatory evidence that this heat transfer coefficient is indeed equal to $0.85065 \times 10^{-9} \text{ WK}^{-4}$ as given at the end of the previous Section. Evidence pertinent to this conclusion is presented in the present Section as well as Section A.6.

We note in the first place the values of the total excess enthalpy for each day of operation calculated using the "true heat transfer coefficient, $(k_R')_{32} = 0.79350 \times 10^{-9} \text{ WK}^{-4}$ as given by the NHE evaluation as well as those calculated with "true heat transfer coefficient $(k_R')_{262} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ as determined in Section A.4 using the

ICARUS methodology, Table 1. These values are plotted in Figs. A.18 and A.19 respectively. We can see immediately that the evaluation given by NHE must be incorrect because we obtain negative excess enthalpies for some of these days, which contravenes the Second Law of Thermodynamics [c.f. Section A.3]. On the other hand the evaluation based on the heat transfer coefficient given by the modified ICARUS evaluation scheme only gives a very slightly negative excess enthalpy for Day 61.

It is therefore reasonable to assume that the rate of excess enthalpy generation on Day 61 is close to zero. The evaluation of the "lower bound heat transfer coefficient, $(k_R')_{11}$ " from the relevant $(k_R')_{11}$ spreadsheet, Table A.6, must therefore be close to the values of the "true heat transfer coefficient, $(k_R')_{12}$ ". Fig. A.20 gives a plot of the relevant data compared to the plot which we predict using the value $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ and the variation of $(k_R')_{11}$ with time given by the relevant "blank" experiments ^{(4),(13),(14)}. It can be seen that the observed values of $(k_R')_{11}$ are in close accord with those which we would predict on the assumption that there is only a low rate of excess enthalpy generation on that day.

It can be seen that there is only one region of time in which there is a marked deviation from the predicted behavior namely for $0 < t < \sim 10,000 \text{ s}$ on Day 61. In this region $(k_R')_{11}$ is markedly larger than the expected value and, moreover, decreases rapidly with time to these predicted values. It is possible therefore, that the deviation seen in this time range can be attributed to the "overfilling" of the cell [see Footnote C.56]. Separate measurements have shown that the pseudo radiative heat transfer coefficient increases by $\sim 5\%$ over the expected value presumably because of an increase in the conductive contribution through the top of the cell. Another explanation already noted in Section A.2 is the sudden cooling of the cell by the large addition of D_2O on Day 61.

A.6 A Pre-ICARUS Evaluation of the True Heat Transfer Coefficient

It is possible to find a further value of the "true heat transfer coefficient $(k_R')_{12}$ " by applying a method used in 1992 ^{(15),(16)}. It was shown at that time that the "lower bound heat transfer coefficient, $(k_R')_{11}$ " decreases markedly from the expected value during the initial stages of the measurement cycles. In this case the decrease is due to the exothermic absorption of deuterium in the lattice. It would be expected, therefore, that the "lower bound heat transfer coefficient, $(k_R')_{11}$ " would rise markedly to the expected "true" value as this process is completed with the proviso that we can observe a period of operation during which there is zero excess enthalpy generation. It follows that we can derive a value of the "true heat transfer coefficient, $(k_R')_{12}$ " from the maximum of the "lower bound heat transfer coefficient, $(k_R')_{11}$ " which is observed with increasing time.

Fig. A.21 shows the relevant data for the first 57 hours of operation of the experiment [i.e. up to the time of application of the heater calibration pulse on Day 3]. The full line at the top shows the expected variation of $(k_R')_{11}$ with time based on the value of $(k_R')_{12}$ at $t = t_2$ on Day 3 [i.e. $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$], the assumption of zero excess enthalpy generation [i.e. $(k_R')_{11} = (k_R')_{12}$] and the known variation of $(k_R')_{11}$ with time established with "blank" experiments ^{(4),(13),(14)}. It can be seen that $(k_R')_{11}$ does indeed

rise to the predicted levels as the charging of the electrode is completed.

Fig. A.21 also shows the derived rates of excess enthalpy generation based on the experimental values of $(k_R')_{11}$ and the assumption that the "true heat transfer coefficient" is given by the regression line. The horizontal line here gives the expected value of the rate of excess enthalpy generation based on the assumption that the current efficiency for the charging of the electrode is 100% and that the enthalpy of absorption of deuterium in the lattice is 40 kJ Mole^{-1} . It can be seen that the experimental values are in reasonable accord with this prediction.

Fig. A.21 furthermore shows that there is a small build up of excess enthalpy generation on Day 3 following the completion of the charging process [compare ^{(15),(16)}].

The data shown in Fig. A.21 also lead to a number of important questions, which cannot be resolved at the present time [see Footnote C.57].

A.7 Day 68: The Period $0 < t < 21,300 \text{ s}$ During Which the Cell is Driven to Dryness

We consider next the penultimate day of the investigation of the experiment; the cell is driven to dryness during the first part of this measurement cycle. The "raw data" for this section is given in columns 1-5 of Table A.7 while columns 6-9 summarize the interpretation in the relevant NHE spreadsheet [the part using the temperature measurements with the "short thermistor"].

We can draw a number of important conclusions from the "raw data" alone. We note in the first place that the temperature given by the "long thermistor" is now slightly higher than that given by the "short thermistor" whereas the opposite is true for measurements made at low temperatures. At first sight such a change might be attributed to a genuine effect namely, the increase in the enthalpy input in the bottom part of the cell [containing the Pd-B cathode]. However, such an interpretation is unlikely because the temperature difference between the two thermistors is essentially constant for say 20,000 s even though the enthalpy input increases by a factor of three. It is more likely therefore that this particular temperature difference is a further manifestation of errors in the calibration of the thermistors. Experimentally, furious boiling and swirling actions were centered around the cathode during this period which may have affected the temperature readings of the longer thermistor. There was also about 2 cm of foam on the liquid interface.

The temperature differences between the two thermistors are appreciably larger for the last four data acquisition points and this difference is especially marked for the last point [0.590 K]. Such a difference is to be expected if the "long thermistor" is now in the relatively concentrated LiOD solution or foam residue while the "short thermistor" is in the vapor phase. However, we also have to note that the temperature at both positions is above that of the boiling point of pure D_2O . Evidently, we have to take into account the increase of the boiling point with the electrolyte concentration as the D_2O is progressively evaporated [see Section B.2]. However, we also have to take note of the

fact that the vapor phase can be superheated [albeit to only a limited extent, see Footnote C.58]. If we do not take account of the increase of the boiling point with concentration, we arrive at the impossible result of negative enthalpies of evaporation with increasing temperature as shown by the NHE evaluation. We also have to use the correct atmospheric pressure in the calculation of the rate of evaporative cooling, and we need to change the thermoneutral potential and the water equivalent of the cell in the NHE evaluation. As the water equivalent of the cell only leads to a significant term $C_p M (d\Delta\theta/dt)$ in the initial stages for Day 68, it has been assumed that $C_p M$ is unchanged throughout the stage leading to evaporation to dryness [however, see further comments in Section A.8].

The modified spreadsheet is shown in Table A.8, which is still an intermediate step in the evaluation because of the neglect of reflux in the cell. However, this intermediate spreadsheet is sufficient for the next stage of the evaluation. It has already been pointed out in Section B.2 that it is desirable to carry out such a first stage of the evaluation independently of the complexities of the evaluation of the rate of evaporative cooling and reflux in the cell. We can carry out such a first step in a straightforward way by calculating the total enthalpy input and output from the cell using columns 1-3 of Table 9 and the known "true heat transfer coefficient". We obtain:

$$\text{enthalpy input} = 378,105 \text{ J}; C_p M \Delta\theta = 2,012 \text{ J}$$

$$\text{enthalpy output} = 212,686 \text{ J for } (k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$$

$$= 223,320 \text{ J for } (k_R')_{12} = 0.89318 \times 10^{-9} \text{ WK}^{-4}$$

The enthalpy available for evaporation of the D_2O is therefore 163,407 J if the lower value of the heat transfer coefficient applies or 152,773 J if the higher value applies. However, we require 207,031 J for the evaporation of the 4.968 Moles of D_2O in the cell. There is therefore a short fall of either 43,624 or 54,258 J, a short fall, which can only be supplied by excess enthalpy generation in the cell. If this excess enthalpy is supplied at a uniform rate throughout the period leading up to evaporation to dryness, then this would require a mean excess rate of 2.048 or 2.547 W depending on whether the "true heat transfer coefficient" has the lower or higher value.

This calculation is similar to one, which has been described previously ⁽⁸⁾ except that the published version included comments on the time dependence of the rate of excess enthalpy generation. It is quite obvious that the rate of excess enthalpy generation must increase with time because the initial rate on Day 68 is less than 1 W. It is important therefore to try to establish the variation of the rate of excess enthalpy generation with time, if only to make a connection with the initial rate of "Heat after Death" observed after the cell has reached dryness [see Section A.8]. In order to derive this variation, we have to include an estimate of the rate of reflux in the cell, and this part of the calculation will follow the scheme outlined in Section B.2 [see Footnote C.59]. The details of the evaluation are given in Table A.9. We can see that the negative values of the enthalpies seen in Tables A.7 and A.8 are now eliminated as the D_2O in the cell is maintained by the

amount of reflux. The total amount evaporated is also in reasonable accord with the amount of D_2O initially in the cell. It is important to realize that we have assumed that the whole of the heat transfer from the cell in the region filled with vapor leads to recondensation [see Section B.2] i.e. we have overestimated the reflux and underestimated the amount evaporated. We should also note that the calculation is improved somewhat if we allow for the fact that the boiling point reaches a limit due to the limited solubility of $LiOD$ in D_2O at the boiling point [this aspect is not illustrated in this Report].

Although the calculation as outlined gives a reasonable interpretation of the behavior of the cell as the contents are driven to dryness [elimination of negative enthalpies of evaporation], we nevertheless still derive negative rates of excess enthalpy generation at long times. This is undoubtedly due to remaining inaccuracies in the calculation of the rates of evaporative cooling. At the present time it is best to restrict attention to the earlier part of the period leading to evaporation to dryness, say to $t < 18,000$ s. We see from Table A.9 that the rate of excess enthalpy generation reaches ~ 9.3 W at this time, or 27 W cm^{-3} . It is important to realize that similar orders of magnitude are obtained even with the interpretation given by NHE, i.e. the estimate is "robust" [see Footnotes C.60 – 62].

A.8 Day 68: The Period $21,300 \text{ s} < t < 86,400 \text{ s}$ Following Evaporation to Dryness

As has been noted in Section A.1 one of the objectives of the present investigation has been the search for the presence [or absence] of the effects of "Heat after Death". The period following the evaporation to dryness on day 68 is an example of the protocol originally described as Case (5)^{(9),(10)} i.e., cell empty: cell allowed to boil dry; cell then maintained at the rail voltage of the galvanostat.

The original investigation was divided into two parts:

- (i) the investigation and interpretation of the cooling curves following evaporation to dryness;
- (ii) the evaluation of thermal balances in the corresponding period.

Attention here will be confined to the second of these approaches.

Table A.10 gives the "raw data" for the first 9000 s of operation during this period [columns 1 – 4]. The data set has been restricted to the first 30 acquisition intervals so as to avoid the complications due to the heater calibration pulse, which was still applied on Days 68 and 69. Columns 5 and 6 gives the data required for the calculation of the rates of excess enthalpy generation shown in columns 7 and 8. It has been assumed that the water equivalent of the empty cell is 28.3 JK^{-1} , the value for the cell filled with electrolyte less the contribution due to the 5 moles of D_2O removed during evaporation to dryness.

The values of the rates of excess enthalpy generation shown in column 7 have been based

on the "true heat transfer coefficient, $(k_R')_{12}$ " observed for the cell filled with electrolyte, i.e. $0.85065 \times 10^{-9} \text{ WK}^{-4}$, which will certainly apply to initial stage of the observation of "Heat after Death" when the cell is filled mainly with D_2O vapor. However, calibrations of cells filled with air ^{(9),(10)} have shown that the heat transfer coefficient falls to about 0.75 of the value for the cells filled with electrolyte. The values of the rates of excess enthalpy generation shown in column 8 have therefore been calculated using $(k_R')_{12} = 0.65 \times 10^{-9} \text{ WK}^{-4}$.

As would be expected, the rates of excess enthalpy generation shown in column 8 are lower than those in column 7, but the decreases in both cases are broadly in line with those expected for a process controlled by a diffusional relaxation time. The initial rate of excess enthalpy generation given by column 7 is approximately the same as the rate reached during the period $0 < t < 21,300 \text{ s}$ as the cell is being driven to dryness, Fig. A.22. Such a correspondence would, of course, be expected if excess enthalpy generation takes place in the bulk of the metal phase.

We note also that the rate of excess enthalpy generation is about 10 times that of the rate of enthalpy input during this period of "Heat after Death".

A.9 Day 69: The Period $2400 \text{ s} < t < 32,400 \text{ s}$

This period is of special interest in the operation of the cell because the cell was disconnected from the galvanostat at 2400 s so that the enthalpy input was zero during the remaining period of operation. In any search for the effects of "Heat after Death" the protocol there should be described as Case (6) ^{(9),(10)} i.e. cell empty: cell allowed to boil dry; cell disconnected from the galvanostat and that the application of Case (6) was preceded by a period covered by Case (5) as described in Section A.8.

The examination of the behavior of the cell has been restricted here to the time $t < 32,400 \text{ s}$ as the usual calibration pulse was applied at $t_1 = 32,400 \text{ s}$. The Joule heat injected by the calibration system is developed in a small volume so that this calibration cannot be used to derive the "true heat transfer coefficient" of the cell for the particular operating conditions [see Footnote C.63].

The "raw data" for this period of operation are given in Table A.11 and a "cooling curve" is plotted in Fig. A.23. It can be seen that although the temperature differences between the cell and water bath are small, they are nevertheless significant. Inspection of Fig. A.23 shows that there must be a source of enthalpy in the system firstly, because the rate of cooling at short times is too slow to be accounted for by the cooling of a calorimeter with a water equivalent of 28.3 JK^{-1} and any conceivable value of the heat transfer coefficient; secondly, because we can detect at least one period during which the cell contents are reheated!

The data in Table A.11 are suitable for the analysis of the cooling curve according to the method originally outlined ^{(9),(10)} using the equation

$$\begin{aligned} \ln[y_0(1+y)/y(1+y_0)] + \tan^{-1}(1+y) - \tan^{-1}(1+y_0) \\ = 4(k_R')\theta_{\text{bath}}^3 t/C_p M \end{aligned} \quad (\text{A.3})$$

$$y = \Delta\theta/\theta_{\text{bath}} \text{ and } y_0 = \Delta\theta_0/\theta_{\text{bath}} \quad (\text{A.4})$$

and $\Delta\theta_0$ is the initial temperature difference.

Fig. A.24 shows a plot of the experimental data according to equation (A.3); the full line shows the predicted behavior using $C_p M = 28.3 \text{ JK}^{-1}$ and $(k_R') = 0.65 \times 10^{-9} \text{ WK}^{-4}$. The deviations from this plot due to enthalpy generation are similar to those previously observed ^{(9),(10)}.

We can also make thermal balances at each point of the cooling curves using particular values of the water equivalent and "true heat transfer coefficient". Those based on $C_p M = 28.3 \text{ JK}^{-1}$ and $(k_R')_{12} = 0.65 \times 10^{-9} \text{ WK}^{-4}$ give initial rates of enthalpy generation $\sim 0.5 \text{ W}$. Unfortunately, the thermal balances in the period preceding the disconnection of the cell from the galvanostat [i.e. the last part of Case 5, Section A.8] cannot be made with sufficient accuracy to allow a comparison of the rates of enthalpy generation at the end of the period following Case 5 and the beginning of the period following Case 6 [see comparison of the rates at the end of the period leading to evaporation to dryness and the beginning of the period following Case 5, Sections A.7 and A.8].

A.10 Days 25 and 26: The Period Day 25 + 76,300s < t < Day 26 + 22,300s

As has already been noted in Section A.2, there were frequent changes of current density in the experiment. Consideration of Case 1 of the conditions likely to give demonstrations of the phenomenon of "Heat after Death" ^{(9),(10)}:

Cell full: cell operated at intermediate temperatures; cell current then reduced in stages [Case 1]

shows that the change of current close to the start of Day 26 of the measurement cycles is likely to provide the best example of this particular case, see Fig. A.2. There are two principal reasons, which indicate this. In the first place, the current density at the end of Day 25 is above the threshold value required for the observation of the phenomenon ⁽¹²⁾ while on Day 26 it is below this threshold value. Secondly, the cell temperature on Day 25 is above that which has been observed to be important for the onset of "positive feedback" ^{(8),(15),(16)} whereas on Day 26 it drops below this value. We would therefore expect to see a marked decrease of the rate of excess enthalpy generation at the start of Day 26 from the value, which applies at the end of Day 25 to that which applies towards the end of Day 26 [see Footnote C.64].

The data covering measurements in the last stages of Day 25 and the beginning of Day 26 are given in Table A.12. Here columns 1-4 give the "raw data", columns 5 - 8 give the derived quantities used to define the "lower bound heat transfer coefficient, $(k_R')_{11}$ " shown in column 9. We note that we used the value of $C_p M = 475 \text{ JK}^{-1}$ in view of the

overfilling of the cell on Day 26 [see Section A.2]. The rates of excess enthalpy generation derived are plotted in Fig. A.25. We can see the well-defined fall in the rate of excess enthalpy generation, which, as in the other examples of Heat after Death discussed in this Report, is consistent with a diffusional relaxation time.

Columns 11 and 12 of Table A.12 also give the "lower bound heat transfer coefficient, $(k_R')_{11}$ " and rate of excess enthalpy generation taken from the spreadsheets for the evaluations carried out by NHE. We can see both from column 13 of this Table and from the plot in Fig. A.26, that this evaluation predicts a negative rate of excess enthalpy generation on Day 25. As we have noted elsewhere in this Report, such negative rates violate the Second Law of Thermodynamics and are certainly due to the use of the incorrect value of the "true heat transfer coefficient, $(k_R')_{12}$ " given by the NHE analysis. Nevertheless, we can see from Table 13, or Fig. A.26, that we can detect the effects of "Heat after Death" on Day 26 even when using this faulty analysis. Furthermore, the increasing values of the "lower bound heat transfer coefficient $(k_R')_{11}$ " on that day demonstrate the presence of a rate of excess enthalpy generation, which decreases with time.

It is important, however, to draw attention to a remaining difficulty in the interpretation, namely, that the initial rate of excess enthalpy generation on Day 26 is larger than the final rate on Day 25. The discrepancy would be diminished if the water equivalent were even higher than 475 JK^{-1} or if we increased $(k_R')_{12}$ in view of the evident increase in the D_2O content of the cell, Fig. A.4. A higher water equivalent is not likely based on the experimental measurements of the electrolyte volume. It does not seem possible, therefore, to eliminate the effect by any sensible choice of the values of $C_p M$ and $(k_R')_{12}$ thus the effect is likely to be real. If this is so, then the establishment of "Heat after Death" and/or "positive feedback" would be more complicated than is indicated by the state variables alone. For example, the time derivatives may also be involved ^{(6),(7)}. It is evident that much further work is required on these particular aspects. This work would be justified not only from the objective of clarifying the science involved but, also, because the judicious use of "positive feedback" and "Heat after Death" offers us the prospect both of increasing the power density and at the same time, of increasing the energy efficiency. It should be noted that if we exclude the enthalpy input due to the cooling of the cell, the rate of excess enthalpy generation in the initial stages of Day 26 is approximately equal to the enthalpy input, i.e. a power gain of $\sim 100\%$ whereas it approaches $\sim 1000\%$ for the initial stages of Heat after Death according to Case 5 ^{(9),(10)}, Section A.8, and infinity % for the example of Case 6, Section A.9. It appears that if the cooling of such cells is prevented [effectively by raising the temperature of the heat sink], then enthalpy generation may be maintained for long durations [~ 1 week] at very high energy efficiencies ⁽²⁶⁾. It is evident that this aspect of the work requires extensive further investigation particularly with regard to attempts to raise the power density of such devices while maintaining the high energy efficiency.

A.11 Further Comments and Conclusions

This Pd-B experiment exhibits all the key features, which have been found in earlier

work. These are in the main:

- (i) excess enthalpy generation in the early stages [$t < 2$ Days] due to absorption of deuterium in the lattice followed by
- (ii) a build up of the rate of excess enthalpy generation
- (iii) the development of "positive feedback" i.e. the increase in the rate of excess enthalpy generation with increase of temperature
- (iv) a marked increase in the rate of excess enthalpy generation at temperatures close to the boiling point of the electrolyte
- (v) a variety of examples of the phenomenon of "Heat-after-Death" i.e. a maintenance of elevated rates of excess enthalpy production following reduction of the current density which may be accompanied by the complete evaporation of the electrolyte.

At the same time there are some marked differences between this experiment using Pd-B and the earlier investigations: the effects of some of these differences can be explained in terms of the earlier results while some of the results are surprising. The major difference is that the measurement cycles had to be carried out at rather low current densities [low for the observation of the phenomenon] in view of the relatively high surface area of the electrode [it is necessary to limit the power input to the cell to satisfy the design criteria of the calorimeter]. As the rate of excess enthalpy generation increases markedly with the current density ⁽¹²⁾, the values achieved in the experiment were necessarily limited [the rates increased to $\sim 27 \text{ W cm}^{-3}$ on Day 68 prior to evaporation to dryness]. A secondary consequence of the low current densities was that the electrode was polarized in the vicinity of the region for the onset of "positive feedback" for most of the experiment duration [see Fig. A.2]. The use of such conditions is known to limit the rates of excess enthalpy generation, and, in the limit, may destroy the phenomenon ^{(6),(7)} [see Footnote C.65].

The major unexpected difference for this Pd-B electrode has been the observation of the development of "positive feedback" at a very early stage of the experiments [Day 3], at a low current density and at a low temperature. It is obviously very important to establish whether this early establishment of "positive feedback" is a property of Pd-B alloys [the one used in this study] or whether some other factor is involved. The marked increase in the rate of excess enthalpy generation at temperatures approaching the boiling point is probably due to non-uniform charging [see Footnote C.66]

A major feature of the investigation of "Heat-after-Death" in the experiment is the demonstration that the rates of excess enthalpy generation before and after the onset of the phenomenon are probably identical. Such an identity would be expected if excess enthalpy generation takes place in the bulk of the electrode, but these effects clearly require further investigation. It is also apparent that these processes relax with a

diffusional relaxation time and prolonged maintenance of the effects evidently requires special conditions ⁽²⁶⁾ [increase of the temperature of the heat sinks]. Results for Day 25 and Day 26 shown in Figure A-25 also indicate new complications in the establishment of "Heat-after-Death" and/or "Positive Feedback".

This investigation of this Pd-B experiment has demonstrated yet again that certain methods of data evaluation are unsound and/or inaccurate or imprecise [compare e.g. (4),(5),(6),(7),(13),(14)]. Furthermore, it is essential to avoid the effects of "positive feedback" as it is impossible in general to calibrate closed loop systems subject to such feedback. Calibrations can only be achieved if the effects are not too marked, as has been the case for Day 3 of this study. Unfortunately, it is almost certain that the investigations carried out by NHE have relied precisely on such unsound and inaccurate methods of calibration and the effects of "positive feedback" have been ignored. However, this neglect is probably quite general and, no doubt, accounts for many of the contradictory results in this field of research. It should be noted that much of the pointless controversy in this field could have been avoided if it had been possible to replace the ICARUS 1 - 3 Calorimeters, Fig. A.1, by the ICARUS-4 version [later reclassified as the ICARUS-14 Calorimeter] Fig. A.27. While it is not certain that this particular redesign would have eliminated the weak time-dependence of the heat transfer coefficients observed with the ICARUS-1 Calorimeter, it is likely that this would have been true and that these systems could have been developed so that all measurements could have been evaluated with a single, predetermined value of the "true heat transfer coefficient". Finally, it is important to note that it has been possible to achieve:

(vi) a satisfactory interpretation of evaporation to dryness [Day 68]

This interpretation has had to take into account: the actual barometric pressure, the change of the boiling point of the solution with increasing electrolyte concentration, [saturation of the electrolyte - not discussed in the present Report], changes in the reflux ratio [see Footnote C.67]. However, prolonged investigations of boiling conditions ^{(17),(27)} will clearly require the design and application of dual calorimeters such as the ICARUS-9 version ^{(17),(27)}. It is also important to determine whether the marked increase of the rates of excess enthalpy generation at temperatures near the boiling point are dependent on the establishment of boiling conditions or are simply due to the increase in temperature. While it is certainly desirable to develop pressurized systems to increase the boiling point, significant increases in the boiling point could also be achieved by using concentrated electrolyte solutions. The use of such electrolytes would allow the extension of the range of applicability of the ICARUS-1 Calorimeters.

Finally, we can note that the interpretation of this experiment gives a good illustration of the need to evaluate all such measurements as individual "Case Histories": the state of development of research in this field in 1993 [when the first ICARUS System was constructed] was certainly not at the point at which such interpretations could be carried out as a matter of routine. Furthermore, the instrumentation also required a number of additional developments to facilitate any such attempts at routine evaluations. The ICARUS-14 System [then described with the label ICARUS-4] was to be the next step but, as has already been noted, this modification could not be accomplished.

Section B

B.1 Introduction

This Section contains comments on the modeling of the Isoperibolic Calorimeters used in the ICARUS-1 and -2 Systems leading to the definition of a number of versions of the heat transfer coefficient, which we have found useful in our investigations [these heat transfer coefficients define the behavior of the calorimetric systems]. The content of this Section is based on Items 16 - 18 of an earlier Document ⁽¹³⁾ [see Footnote C.6] and which should be read in conjunction with the present Report. The earlier Document ⁽¹³⁾ also contained examples of data evaluations using both simulated data and results for "blank" experiments [with Pt-cathodes; compare ⁽¹⁴⁾]. These illustrations ⁽¹³⁾ demonstrated the reasons and validity of the choice of the restricted set of heat transfer coefficients specified for the application of the ICARUS-1 Methodology ⁽¹²⁾.

B.2 Modeling of the Calorimeters

It has been established that at low to intermediate cell temperatures [say $30^\circ < \theta < 80^\circ\text{C}$] the behavior of the calorimeters is modeled adequately by the differential equation:

$$\begin{aligned}
 C_p M (d\Delta\theta/dt) &= [E_{\text{cell}}(t) - E_{\text{thermoneutral, bath}}] I + Q_r(t) \\
 \text{change in the} & \quad \text{enthalpy input} & \quad \text{rate of excess} \\
 \text{enthalpy content} & \quad \text{due to} & \quad \text{enthalpy} \\
 \text{of the calorimeter} & \quad \text{electrolysis} & \quad \text{generation} \\
 + \Delta Q_H(t-t_1) - \Delta Q_H(t-t_2) - (3I/4F)[P/\{P^* - P\}][C_{p, D_2O, g} - C_{p, D_2O, l})\Delta\theta + L] \\
 \text{calibration pulse} & \quad \text{rate of enthalpy removal by the gas stream with} \\
 & \quad E_{\text{thermoneutral}} \text{ referred to the bath temperature} \\
 - (k_R o) \theta_{\text{bath}}^3 [1 - \gamma t] \{f_1(\theta)/\theta_{\text{bath}}^3 + 4\phi\Delta\theta\} \\
 \text{time dependent} & \quad \text{effect of} & \quad \text{effect of} \\
 \text{heat transfer} & \quad \text{radiation} & \quad \text{conduction} \\
 \text{coefficient.} & & \\
 \end{aligned} \tag{B.1}$$

With the calorimeters supplied with the ICARUS Systems, the conductive contribution to heat transfer was very small. In fact, if this term was "lumped" into the radiative term by allowing for a small increase in the radiative heat transfer coefficient:

$$\text{Radiative heat transfer} = (k_R' o)[1 - \gamma t][(\theta_{\text{bath}} + \Delta\theta)^4 - \theta_{\text{bath}}^4] \tag{B.2}$$

then, the values of the "pseudo-radiative" heat transfer coefficient derived, $(k_R' o)[1 - \gamma t]$, were close to those calculated from the Stefan-Boltzmann coefficient and the radiative

surface area [typical values $0.72 \times 10^{-9} \text{ WK}^{-4} < (k_R' o) [1 - \gamma] < 0.76 \times 10^{-9} \text{ WK}^{-4}$]. However, for the Cell No.38 used in this experiment this "pseudo-radiative" heat transfer coefficient is found to be $\sim 0.85 \times 10^{-9} \text{ WK}^{-4}$ so that the conductive contribution was evidently increased. We have to assume that this increase in the heat transfer coefficient must have been due to a "softening" of the vacuum in the Dewar calorimeters while sitting in the laboratory for several years.

An increase in the "pseudo-radiative" heat transfer coefficient can normally only be observed if the cells are "overfilled" with D_2O during the periodic replenishment of the cells. This "overfilling" of the cells leads to the approach of the electrolyte level to the base of the Kel-F plug sealing the cells thereby increasing the conductive losses through the top of the cell. This effect leads to a 4 - 5% increase in the "pseudo-radiative" heat transfer coefficient as possibly observed in the results for Day 61 of this experiment [see Figure A.20, Section A.5 and Footnote D.4].

If the time dependence of the heat transfer coefficient is not included explicitly in equation (B.2) then:

$$\text{Radiative heat transfer} = (k_R') [(\theta_{\text{bath}} + \Delta\theta)^4 - \theta_{\text{bath}}^4] \quad (\text{B.3})$$

where the radiative heat transfer coefficient (k_R') now shows a weak time-dependence.

In calculating the rate of enthalpy removal by the gas stream

$$(3I/4F)[P/\{P^* - P\}][(C_{P,D_2O,g} - C_{P,D_2O,l}) \Delta\theta + L] \quad (\text{B.4})$$

we have always assumed that the partial pressure of D_2O [or H_2O] in this gas stream can be calculated using the Clausius-Clapeyron equation with the latent heat of evaporation, L , being that at the boiling point. Evaporative cooling only becomes an important term at temperatures close to the boiling point [say, at $\Delta\theta > 70^\circ\text{C}$] where these two assumptions are justified. At low to intermediate temperatures, $\Delta H_{\text{evap}}(t)$ is a minor correction term so that errors due to the two assumptions introduce second order small quantities. In particular the errors introduced by neglect of the temperature dependence of the latent enthalpy of evaporation are $< 0.1\%$ under all conditions of operation of the cells. It is also important to bear in mind that such errors are further reduced for all evaluations of the "true" heat transfer coefficients as these evaluations are based on differences in temperature induced by the calibration pulses [or on differences in temperature induced by "topping" up of the cells or perturbations of the current density: such methods of calibration are not considered in this Report].

However, equation (B.4) does not give the rates of enthalpy loss tabulated in the $(k_R')_{11}$ spreadsheets provided by the NHE analyses. The parameters required for this calculation were contained in data files of the ICARUS-1 and ICARUS-2 software and were identical for both systems. The values installed in the programs as supplied were [see Footnote C.8]:

$C_{P,D_2O,l}$	$= 84.349 \text{ J Mol}^{-1} \text{ K}^{-1}$
$C_{P,D_2O,g}$	$= 34.27 \text{ J Mol}^{-1} \text{ K}^{-1}$
θ_{boiling}	$= 374.570 \text{ K}$
F	$= 96484.56 \text{ C Mole}^{-1}$
R	$= 8.314410 \text{ J Mole}^{-1} \text{ K}^{-1}$
L	$= 41,672.600 \text{ J Mole}^{-1}$
P^*	$= 1.003 \text{ Ats}$

The Handbooks ^{(1),(2)} contained specific instructions that some of these parameters would need to be changed [here θ_{boiling} and P^* , see below] as well as instructions as to how such changes in the parameter listing were to be carried out. However, it appears that values of the rates of evaporative enthalpy loss close to those given in the NHE analyses may be calculated for low to intermediate temperatures using the parameter listing supplied with the instruments, i.e. the changes required were not made. [It also appears that the latent enthalpy of evaporation was not corrected for changes in temperature]. The consequent errors are sufficiently small that they do not invalidate the analyses. However, the values of the rates of evaporative enthalpy loss contained in the $(k_R')_{11}$ spreadsheets of the NHE analyses for temperatures close to the boiling point cannot be calculated with any values of the parameters close to those contained in the listing supplied with the instruments. This matter has not been investigated further as it is in any event necessary to make three further changes if experiment cycles close to the boiling point are to be evaluated [see Footnote C.9].

The first of these changes is the adjustment of the boiling point to the value which applies to the ambient atmospheric pressure. It should be noted that the ICARUS-2 System was supplied with the means for the continuous recording of the barometric pressure, but this facility was evidently disabled following the installation of the system at the NHE laboratory. The values of P^* which, have been used in the present interpretation, have been obtained from the Sapporo Airport [thanks to Professor Akito Takahashi of Osaka University], and it has been assumed that the same values apply to the experiments carried out in the NHE Laboratory. Furthermore, it has been assumed that the pressure in the cell is the same as the ambient pressure although it may well be that the pressure in the cell was somewhat higher than this value.

The second change is that it is necessary to take note of the fact that the boiling point corresponds to that of the electrolyte solution in the cell. It has been assumed that this correction is given by that for an ideal solution

$$\Delta\theta_{\text{boiling point}} = (R/L)(\theta_{\text{boiling point}})^2 \ln X_1 \quad (\text{B.5})$$

where X_1 is the mole fraction of the D_2O in the electrolyte. It will be evident that this correction becomes especially important on Day 68 when the cell contents are driven to dryness. In that case the boiling point must be adjusted at each measurement interval as the D_2O content of the cell decreases. The values of the boiling points appropriate for the interpretation of the experimental data for Day 68 are discussed in Section A.7.

The third change again applies specifically to Day 68, namely, an allowance for the effect of reflux in the cell. In order to evaluate the effects of reflux we need to take note of the fact that the vapor space in the cell is filled predominantly by D₂O as the cell is driven to dryness. Thus, even at the start of Day 68 the mole fraction of D₂O in the vapor space was ~ 0.85 for this experiment. In consequence, heat transfer from the vapor phase to the walls of the Dewar [to provide the enthalpy input required by radiation across the vacuum gap] was dominantly from the D₂O content of the vapor. We also need to take note of the fact that the contribution to the heat capacitance of the vapor phase in the vicinity of the boiling point due to the D₂O content of this phase is

$$-d(LP/P^*)/d\Delta\theta = L^2/R(\theta_{\text{boiling point}})^2 \exp[-L\Delta\theta/R(\theta_{\text{boiling point}})^2] \quad (\text{B.6})$$

where $\Delta\theta$ is the temperature displacement from the boiling point. This heat capacitance is ~ 67 times larger than that of an equivalent gas space filled with oxygen and hydrogen and therefore ~ 380 times larger than the heat capacitance due to these gases for the actual working conditions at the start of day 68.

This marked increase in the heat capacitance of the part of the cell filled with gas and D₂O vapor has two consequences. In the first place, the heat transfer across the vacuum gap must be maintained at the same value as that which applies to the liquid phase [see Footnote C.10]. Secondly, the radiative output across the section of the Dewar cells filled with vapor must be balanced by the condensation of an amount of vapor sufficient to supply the radiative enthalpy. A first approximation for the reflux in the cell is therefore,

$$\text{Rate of reflux} = (k_R')_{12} f_1(\theta) \Sigma \Delta M / L \Delta t M^0 \quad (\text{B.7})$$

where ΔM is amount of D₂O evaporated in each measurement interval Δt .

Equation (B.7) will be an upper limit for this extent of reflux since we are neglecting the heat transfer to the walls by the deuterium and oxygen in the gas space as well as the effects of the re-heating of this gas space by the liquid in the lower section of the calorimeter [see Footnotes C.11, D.5, E.2].

It will be evident that analyses based on the use of equations (B.4) - (B.7) can only be approximations for a number of reasons in addition to those which have been delineated. Two of the most obvious deficiencies are the use of "dilute solution theory" in the interpretation and the neglect of hydrostatic pressure on the boiling points used in the Clausius-Clapeyron equation. It follows, therefore, that a part of the analyses of the "raw data" for the episodes of cells being driven to dryness should be based on assumptions which are independent of the use of equations (B.4) - (B.7). These matters are considered further in Section A.7.

B.3 Definition of the Heat Transfer Coefficients Used in the ICARUS Data Analyses.

The heat transfer coefficients will be described by the suffices used previously ⁽¹³⁾ [as well as in earlier Reports and Correspondence] as defined in Section A.2

$$(k_R')_{ij,l} \quad (B.8)$$

where $i = 1, 2, 3$ denotes "differential", "backward integration" and "forward integration", j is defined at appropriate points below [see also Section A.2] and $l = 1, 2$ denotes "lower bound" and "true" respectively [see Footnote C.12].

The simplest starting point is to assume that there is no excess enthalpy generation in the system i.e.

$$Q_f(t) = 0 \quad (B.9)$$

in equation (B.1) and to evaluate a "lower bound heat transfer coefficient" [i.e. a coefficient which assumes that the rate of excess enthalpy generation is zero] at a time just before the end of the calibration pulse, $t = t_2$:

$$(k_R')_l = [(E_{\text{cell}}(t) - E_{\text{thermoneutral, bath}})I - \Delta H_{\text{evap}}(t) - C_p M(d\Delta\theta/dt)]/f_l(\theta) \quad (B.10)$$

This was the first heat transfer coefficient used in our investigations, hence the designation $(k_R')_l$. It should be noted that this designation should really be changed so as to be consistent with the definition (B.8), but this will not be done principally because the definition (B.10) was subsequently extended to any part of the measurement cycle, the coefficient being designated $(k_R')_{11}$. We should perhaps change this designation of $(k_R')_{101}$ to denote

$$\begin{aligned} i &= 1, \text{ differential} \\ j &= 0, \text{ any part of the measurement cycle} \\ l &= 1, \text{ lower bound} \end{aligned}$$

but the description $(k_R')_{11}$ will be retained as it has been used extensively in earlier reports and papers.

Having obtained $(k_R')_{11}$, it is frequently desirable to establish the 11-point averages $(\overline{k_R'})_{11}$ so as to decrease the "noise" [see Footnote C.13, D.6 and E.3]. Such averaging gives ~ 26 independent values for measurement cycles lasting 1 day, or better ~ 52 values for the recommended 2-day cycles. In turn, it is useful to evaluate the 6-point averages of $(\overline{k_R'})_{11}$ which have been designated as $(\overline{\overline{k_R'}})_{11}$. It is not useful to extend this averaging beyond 6 points because any such extension makes the systematic errors [due to the residual decrease of $(k_R')_{11}$ with time] larger than the random errors.

The values of $(\overline{k_R'})_{11}$ and $(\overline{\overline{k_R'}})_{11}$ [and the relevant statistics] were to have been included in the " $(k_R')_{11}$ - spreadsheets" but it is apparent that the group at NHE did not

carry out these averaging procedures.

It is apparent from equation (B.10) that we need accurate values of $C_p M$ to make $(k_R')_{11}$ generally useful, but it is also apparent that the group at NHE retained the value of $C_p M$ specified in the parameter listing rather than to determine the correct value and to substitute this corrected value in the listing [see Footnote C.14]. A first approach to the determination of the value of $C_p M$ for any given cell is to rearrange (B.10) to the straight line form

$$y = mx + c \quad (B.11)$$

i.e.

$$\begin{aligned} & [\{E_{\text{cell}}(t) - E_{\text{thermoneutral, bath}}\} I - \Delta H_{\text{evap}}(t)] / f_1(\theta) = \\ & [C_p M (d\Delta\theta/dt)] / f_1(\theta) + (k_R')_{1,j,l} \end{aligned} \quad (B.12)$$

and to derive then approximate values of $C_p M$ from the slopes of the plots in regions where the temperature is varying relatively rapidly with time. We can distinguish four such plots designated by the relevant derived heat transfer coefficients $(k_R'o)_{151}$, $(k_R'o)_{161}$, $(k_R'o)_{171}$ and $(k_R'o)_{181}$ according to whether the fitting of equation (B.12) is carried out at times somewhat above the origin, at times somewhat above t_1 [the time of application of the calibration pulse], at times somewhat above t_2 [the time of cessation of the calibration pulse] or by the combination of the last two time regions, see Fig. A.11. However, there is a measure of ambiguity about the interpretation of the values of $(k_R'o)_{1,j,l}$ derived which has been discussed in Items 20 and 21 of ⁽¹³⁾.

It should be noted that $(k_R'o)_{151}$ cannot be evaluated systematically for this Pd-B experiment because of the irregular schedule of the addition of D_2O , see Section A.2. Evaluations of $(k_R'o)_{161}$, $(k_R'o)_{171}$ and $(k_R'o)_{181}$ for the important data set for Day 3 are markedly degraded due to the early onset of "positive feedback", see Section A.3. The procedure based on equation (B.12) has limited precision because of the need to differentiate the inherently "noisy" experimental data. It is therefore necessary to carry out the fitting procedures over extended regions of the abscissa, $(d\Delta\theta/dt)/f_1(\theta)$, so that the data are inevitably affected by the onset of the "positive feedback" detected for the operation of the cell on that day.

In this connection it should also be noted that separate investigations have shown that $(d\Delta\theta/dt)$ is best estimated by using the second order central differences [i.e. the chords of the curves]. More accurate values could be derived in principle by using higher order differences. However, in practice, the repeated differentiation of the experimental data [implicit when using higher order differences] leads to an increase in "noise" if we use differences higher than the second order [see also Footnote C.15].

In the absence of sufficiently precise determinations of $C_p M$, the evaluations must necessarily be restricted to regions of time where the contribution of the term $C_p M (d\Delta\theta/dt)/f_1(\theta)$ is adequately small. In that case it is adequate to use a "guesstimate" of $C_p M$. This matter [including the evaluation of a "guesstimate" of $C_p M$] is considered

further in Section A.3.

It is next necessary to evaluate a "true differential heat transfer coefficient". The simplest procedure, giving $(k_R')_2$ near the end of the calibration period at time $t = t_2$, is obtained by including the calibration pulse, ΔQ :

$$(k_R')_2 = \{ \Delta Q + [E_{\text{cell}}(\Delta\theta_2, t_2) - E_{\text{cell}}(\Delta\theta_1, t_2)]I - \Delta H_{\text{evap}}(\Delta\theta_2, t_2) + \Delta H_{\text{cvap}}(\Delta\theta_1, t_2) - C_p M [(d\Delta\theta/dt)_{\Delta\theta_2, t_2} - (d\Delta\theta/dt)_{\Delta\theta_1, t_2}] \} / f_2(\theta) \quad (\text{B.13})$$

where we now have

$$f_2(\theta) = [\theta_{\text{bath}} + (\Delta\theta_2, t_2)]^4 - [\theta_{\text{bath}} + (\Delta\theta_1, t_2)]^4 \quad (\text{B.14})$$

$[(k_R')_2$ was the second heat transfer coefficient used in our investigations]. In order to carry out such evaluations, it is useful to construct A.4 or A.3 sized plots of the "raw data" and then to obtain appropriate averages by using a transparent ruler. This type of analysis used to be a generally accepted approach but then fell into disrepute. However, the methodology is now again accepted giving so-called "robust estimates".

It may be noted that the errors in $(k_R')_2$ are measures of the accuracy of the "true heat transfer coefficient" as the estimates are made in terms of the known Joule enthalpy input to the calibration heater. Errors in $(k_R')_1$ or $(k_R')_{11}$ are measures of the precision of the "lower bound heat transfer coefficients" as there is no independent calibration and there may be excess enthalpy generation in the system. It is important that $(k_R')_1$ and $(k_R')_2$ are the least precise and least accurate coefficients, which can be obtained from the "raw data". Statements that the errors are larger than this [e.g. see ⁽¹¹⁾] simply show that mistakes have been made in the data analysis procedures and/or the execution of the experiments.

We have always insisted that the construction and evaluation of plots of the "raw data" is an essential prerequisite of the more elaborate data evaluation procedures. For one thing it shows whether the "noise levels" in the experiments were sufficiently low to justify more detailed evaluations and also points to malfunctions in the experiments. It also shows immediately whether the θ - t and E_{cell} - t transients have relaxed sufficiently to permit the evaluation of $(k_R')_1$ and $(k_R')_2$. Furthermore, it gives immediate indications of the presence [or absence] of "positive feedback". As has been pointed out repeatedly, all calibration procedures require that the rate of excess enthalpy generation, $Q_i(t)$, be constant during the calibration periods. These matters are considered further in the main text, Section A.3 and A.4 [see Footnote E.5].

Having obtained the "true heat transfer coefficient" at a single point [usually near the end of the calibration pulse, $t = t_2$] it is important to ask: "what is the true heat transfer coefficient, $(k_R')_{12}$, at any other time?" We can make such an evaluation within the duration $t_1 < t < t_2$ of the calibration pulse simply by using equation (B.13) giving $(k_R')_{12}$ rather than $(k_R')_2$. Note also that equation (B.13) can be rearranged to the straight line

form:

$$\begin{aligned} & \{\Delta Q + [E_{\text{cell}}(\Delta\theta_2, t) - E_{\text{cell}}^0(\Delta\theta_1, t)]I - \Delta H_{\text{evap}}(\Delta\theta_2, t) + \Delta H_{\text{evap}}(\Delta\theta_1, t)\} / f_2(\theta) \\ & = \{C_p M [(d\Delta\theta/dt)_{\Delta\theta_2, t} - (d\Delta\theta/dt)_{\Delta\theta_1, t}]\} / f_2(\theta) + (k_R'o)_{162} \end{aligned} \quad (\text{B.15})$$

which is applicable at times close to and above t_1 . It is evident, therefore, that such plots can also be used to obtain estimates of $C_p M$, but the accuracy of such values is inevitably much lower than the precision of those obtained by the application of the corresponding expression for the "lower bound heat transfer coefficient, $(k_R'o)_{161}$ " equation (B.12). Nevertheless, equation (B.15) is useful because it allows the removal of the effects of the water equivalent, $C_p M$, on the "true heat transfer coefficient, $(k_R'o)_{162}$ ", simply by extrapolating to zero value of the abscissa. However, the time corresponding to this point will not be accessible experimentally for calibrations carried out with a calibration pulse of 6-hour duration for polarizations carried out at low cell currents [although this time is probably close to $t = t_2$, see Footnote C.16].

In the regions in which there is no application of a heater pulse i.e. for $0 < t < t_1$ and $t_2 < t < T$, the "true heat transfer coefficient" can only be obtained from the "heating" and "cooling curves" i.e. the "driving force" is the change in the enthalpy content of the calorimeters rather than ΔQ . It is now sensible to cast equation (B.13) in the form:

$$\begin{aligned} & \{C_p M [(d\Delta\theta/dt)_{\Delta\theta_2, t} - (d\Delta\theta/dt)_{\Delta\theta_1, t}]\} / f_2(\theta) = - (k_R'o)_{152} \text{ or } (k_R'o)_{172} + \\ & \{[E_{\text{cell}}(\Delta\theta_2, t) - E_{\text{cell}}(\Delta\theta_1, t)]I - \Delta H_{\text{evap}}(\Delta\theta_2, t) + \Delta H_{\text{evap}}(\Delta\theta_1, t)\} / f_2(\theta) \end{aligned} \quad (\text{B.16})$$

If the system is functioning correctly, then it will be found that the L.H.S. of equation (B.16) is essentially constant [although this constancy can only be probed over a short time range]. The second term on the R.H.S. of equation (B.16) will be much smaller than the term on the L.H.S., i.e. it is in the nature of a correction term to give "point-by-point" values of $(k_R'o)_{152}$ or $(k_R'o)_{172}$. It will be evident that the accuracy of these versions of the "true heat transfer coefficient" is limited by the accuracy of the estimates of $C_p M$. This particular part of the methodology is therefore only useful to serve as a check on the operation of the cells and methods of data evaluation. Furthermore, it is not possible to apply equation (B.16) systematically to the time region $0 < t < t_1$ for this experiment in view of the irregular schedule of addition of D_2O to the cell.

As has been noted previously ⁽¹³⁾, we have been unable to combine data in the regions just above t_1 and t_2 to give a simple equation leading to $(k_R'o)_{182}$.

The assumption underlying this part of the account presented in this Report is that we can only determine $(k_R')_{12}$ within the duration of the calibration pulse $t_1 < t < t_2$, and, at a lower accuracy, $(k_R'o)_{152}$ and $(k_R'o)_{172}$ in regions adjacent to the origin and for times adjacent and above t_2 respectively. However, this conclusion is incorrect. We need to make the additional assumption that the rate of any excess enthalpy generation is constant during any particular calibration period in order to determine $(k_R')_{12}$. This means that we

can only obtain a single value of this heat transfer coefficient per calibration period and, consequently, a single value of $(k_R')_{12} - (k_R')_{11}$. Two important points follow from this conclusion. In the first place, the precision of $(k_R')_{12}$ must be very nearly equal to the precision of $(k_R')_{11}$. Secondly, and related to the first point, we see that if we extend the assumption that the rate of excess enthalpy production is constant during the period $t_1 < t < t_2$ to saying that it is constant for the whole measurement cycle, $0 < t < T$, then it is immediately possible to derive $(k_R')_{12}$ over the whole of this cycle. Thus, if the difference between the "true" and "lower bound" heat transfer coefficients can be established at any one time [say $\Delta(k_R')_i$ at time t_2] then $[k_R'(t)]_{12}$ at any other time will be given by

$$[(k_R'(t))]_{12} = [(k_R'(t))]_{11} + \Delta(k_R')_{i_2} f_1(\theta)_{i_2} / f_1(\theta)_i \quad (B.17)$$

The ratio $f(\theta)_{i_2} / f_1(\theta)_i$ is of order unity which implies that the shift $(k_R')_{12} - (k_R')_{11}$ is always close to that at the calibration point. Equation (B.17) also points to a further important conclusion. At the time of writing of the Handbook for the ICARUS-1 Systems ⁽²⁾ we believed that the precision of $(k_R')_{12}$ [and of other versions of the "true heat transfer coefficient"] would always be given by the accuracy of that coefficient which is certainly lower than the precision of $(k_R')_{11}$ [see Footnote C.17]. Equation (B.17) shows that is incorrect: the precision of $(k_R')_{12}$ is very nearly equal to the precision of $(k_R')_{11}$. It follows that **changes** in the rates of excess enthalpy production can be established at the same level of precision as that of $(k_R')_{11}$. The same comments apply to the precision of the "true heat transfer coefficient, $(k_R')_{22}$ " relative to that of the "lower bound heat transfer coefficient, $(k_R')_{21}$ " which is discussed below. In consequence, the **changes** in the rates of excess enthalpy production can be established with relative errors $< 0.01\%$, and these errors determine the level of significance with which such changes can be discussed. Of course, the accuracy of the "true heat transfer coefficients" remains determined by the errors of differences such as that of $(k_R')_{12} - (k_R')_{11}$.

It is important here to stress once again that any attempt to calculate the variation of rates of excess enthalpy generation within the measurement cycles must also pay due regard to the fact that it is not possible to calibrate the systems if the rate of excess enthalpy generation varies with time. It is also important that this comment applies equally to any calorimetric system which we might wish to use. If the rate of excess enthalpy generation does, in fact, vary with time, then $\Delta(k_R')$ must be derived from separate experiments. This is the situation, which applies to the experiment as is discussed in Section A.3 and A.4. The comments made in this part of Section B.3 should be read in conjunction with ⁽¹³⁾.

The discussion of the accuracy of "true heat transfer coefficients" versus the precision of the "lower bound heat transfer coefficients" prompted our search for methods, which would increase both the precision and accuracy. The reason for the limited precision of $(k_R')_{11}$ and accuracy of $(k_R')_{12}$ is mainly due to the need to differentiate "noisy" experimental data sets in order to derive $C_p M(d\Delta\theta/dt)$ [see Footnotes C.18 and D.8].

If we wish to avoid the numerical differentiation of the experimental data sets, then we can rely instead on the numerical integrations of these data and compare these to the

integrals of the differential equation representing the model of the calorimeters. For the backward integrals starting from the end of the measurement cycles at $t = T$ we obtain

$$(k'_R)_{21} = \frac{\int_T^t \text{net enthalpy input}(\tau) d\tau}{\int_T^t f_1(\theta) d\tau} - \frac{C_p M [\Delta\theta(t) - \Delta\theta(T)]}{\int_T^t f_1(\theta) d\tau} \quad (\text{B.18})$$

while the corresponding equation for forward integration from the start of the measurement cycle is

$$(k'_R)_{31} = \frac{\int_0^t \text{net enthalpy input}(\tau) d\tau}{\int_0^t f_1(\theta) d\tau} - \frac{C_p M [\Delta\theta(t) - \Delta\theta(0)]}{\int_0^t f_1(\theta) d\tau} \quad (\text{B.19})$$

Here, the suffices 21 and 31 denote respectively "backward integration, lower bound" and "forward integration, lower bound". $(k'_R)_{21}$ and $(k'_R)_{31}$ are the corresponding integral heat transfer coefficients defined at time t .

We have to take note of the fact that care is needed when integrating the terms $f_1(\theta)$ and [net enthalpy input (τ)] around the discontinuities at $t = t_1$ and $t = t_2$ [also the times $t = 0$ and $t = T$ if the range of the integrations is extended]. The trapezium rule, Simpson's rule or the mid-point rule has been used at various times to carry out the integrations. Of these rules, only the mid-point rule is strictly speaking correct in that it agrees with the mathematical definition of an integral. It is quite generally assumed that integrations carried out using the trapezium or Simpson's rule will converge onto the "correct" algebraic result if the integration interval is made adequately small, but this does not necessarily follow. This is a matter which needs to be investigated for each particular case.

It may be noted that the only straightforward way in which we can integrate around the discontinuities at $t = t_1$ and $t = t_2$ is by means of the trapezium rule, and this is the method which has been used in the recalculations presented in this Report. If the times of application and cessation of the heater calibration pulses correspond exactly to t_1 and t_2 , respectively, then we can carry out the integrations around the discontinuities by inserting extra data points at these times. It appears that the data sets in this experiment satisfy this criterion, although this is not generally true for all experiments carried out with the ICARUS-2 system; lack of synchronization of the calibration pulses with t_1 and t_2 appeared to be generally true for measurements with the ICARUS-1 systems. In that case, it is necessary to determine these times separately [this can be done adequately from the $\theta - t$ plots] so as to establish the integration intervals, and it is then necessary to insert 4 additional data points.

The evaluations of $(k'_R)_{21}$ and $(k')_{22}$ [see below] and of $(k'_R)_{31}$ and $(k'_R)_{32}$ [see also below] were to have been carried out using $(k'_R)_{21}$ and $(k'_R)_{31}$ spreadsheets produced by the software. As we have never had access to these spreadsheets [if, in fact, they were ever

produced], we cannot now establish whether the integrations around the discontinuities were carried out correctly, although we believe that they must have been in error. In any event, all the integrations used in the evaluations described in this Report have been carried out using the "raw data".

The adequacy [or inadequacy] of the particular integration procedures coupled to the adequacy of the chosen integration interval is revealed more clearly when we come to the use of equations (B.18) and (B.19) to determine $C_p M$ and to carry out extrapolations to remove the effects of the second term on the R.H.S. of these equations on the corresponding heat transfer coefficients. The procedure set out in the Handbook for the ICARUS-1 System (2) restricted the integrations to the time region of the application of the heater calibration pulse. For backward integration we obtain (B.20)

$$\frac{\int_{t_2}^t \text{net enthalpy input}(\tau) d\tau}{\int_{t_2}^t f_1(\theta) d\tau} = \frac{C_p M [\Delta\theta(t) - \Delta\theta(t_2)]}{\int_{t_2}^t f_1(\theta) d\tau} + (k_R' o)_{261} \quad (\text{B.20})$$

while for forward integration, we obtain

$$\frac{\int_{t_1}^t \text{net enthalpy input}(\tau) d\tau}{\int_{t_1}^t f_1(\theta) d\tau} = \frac{C_p M [\Delta\theta(t) - \Delta\theta(t_1)]}{\int_{t_1}^t f_1(\theta) d\tau} + (k_R' o)_{361} \quad (\text{B.21})$$

It has been found [e.g. see references 3, 4, 5 and 13] that equation (B.20) can be used to derive accurate values of $C_p M$ while there is some minor degradation when using forward integration, equation (B.21). The application of equation (B.20) to the data sets was the "target methodology" of the ICARUS Systems [e.g. see ⁽²⁾] and the derived "lower bound heat transfer coefficient, $(k_R' o)_{261}$ " was described as $(k_R')_{21}$ in the Handbook and the associated correspondence. We have since then used the more extended description, $(k_R' o)_{261}$, to denote the fact that with $j = 6$, we are carrying out the evaluation in the time region $t_1 < t < t_2$. The same types of evaluation may be used to derive $(k_R' o)_{251}$, $(k_R' o)_{271}$ and $(k_R' o)_{281}$ as well as $(k_R' o)_{351}$, $(k_R' o)_{371}$ and $(k_R' o)_{381}$. It is only necessary to start the integrations from the appropriate times, which also give the starting values of θ for the R.H.S. of the relevant equations. Of these sets of estimates, that leading to $(k_R' o)_{281}$ is especially useful and this particular fit also gives good estimates of $C_p M$. However, it should be noted that it is necessary to use care in applying these procedures to the data for Day 3 of this experiment because of the early onset of "positive feedback" see Section A.3.

In order to obtain the "true heat transfer coefficients" it is necessary to combine the

integrals of the enthalpy inputs in equations (B.20) and (B.21) with thermal balances made at one or a series of points. This can be done in a number of ways, and it is important that this part of the evaluation⁽²⁾ was changed in the summer of 1994 following the receipt of the first two sets of data collected by NHE^{(3),(4)}. Attention will be confined here to the procedure originally suggested in the Handbook for the ICARUS-1 System⁽²⁾ [see Footnote C.19]. We make a thermal balance just before the application of the calibration pulse and, if the system has relaxed adequately and $d\theta/dt = 0$, then we consider $(k_R')_{32}$.

$$0 = [\text{Net enthalpy input}(t_1)][t - t_1] + Q_f[t - t_1] - (k_R')_{32} \{ [(\theta_{\text{bath}} + \Delta\theta(t_1))^4 - \theta_{\text{bath}}^4] [t - t_1] \} \quad (\text{B.22})$$

The combination of this equation with equation (B.19) eliminates the unknown rate of excess enthalpy generation. We obtain

$$(k_R')_{32} = \frac{\int_{t_1}^t \text{net enthalpy input}(\tau) d\tau - [\text{net enthalpy input}(t_1)][t - t_1]}{\int_{t_1}^t f_2(\theta) d\tau} - \frac{C_p M [\Delta\theta(t) - \Delta\theta(t_1)]}{\int_{t_1}^t f_2(\theta) d\tau} \quad (\text{B.23})$$

The corresponding equation for $(k_R')_{22}$ follows from equation (B.23) on replacing t_1 by t_2 [see equation (B.24) below].

We note again that the group at NHE did not follow the instruction in the ICARUS-1 Handbook⁽²⁾ to use measurement cycles of 2-day duration and, for the reduced time scales of 1-day cycles in particular, it is necessary to include the term $C_p M (d\Delta\theta/dt)$ in the thermal balances, equation (B.22). However, the team at NHE continued to use the original form of the equation [see Footnote C.20]. It also appears that NHE did not follow the instruction⁽²⁾ to evaluate $(k_R')_{32}$ at times close to t_2 . This matter is discussed further in Section A.4.

It is convenient also to rewrite the derived equation for $(k_R')_{22}$ in the "straight line form".

$$\begin{aligned} & \frac{\int_{t_2}^t \text{net enthalpy input}(\tau) d\tau - [\text{net enthalpy input}(t_2)][t - t_2]}{\int_{t_2}^t f_2(\theta) d\tau} \\ &= \frac{C_p M [\Delta\theta(t) - \Delta\theta(t_2)]}{\int_{t_2}^t f_2(\theta) d\tau} + (k_R')_{262} \end{aligned} \quad (\text{B.24})$$

$(k_R')_{22}$ and $(k_R'o)_{262}$ were the versions of the "true heat transfer coefficient" which we used in our investigations prior to the construction of the ICARUS-1 System. As we did not wish to discuss the differences between $(k_R')_{32}$, $(k_R'o)_{362}$, $(k_R')_{22}$ and $(k_R'o)_{262}$ and, as we expected $(k_R')_{32}$ to converge onto $(k_R')_{22}$ for the specified 2-day measurement cycles [within the error limits specified for the ICARUS-1 System] we also labeled $(k_R')_{32}$ as $(k_R')_{22}$ [see Footnote C.21].

It should be noted that the extrapolation (B.24) automatically removes the effect of the term $C_p M [\theta(t) - \theta(t_2)] / \int_{t_2}^t f_2(\theta) d\tau$ on the "true heat transfer coefficient". This application of equation (B.24) [and of $(k_R')_{22}$ evaluated close to the mid-point $t = t_2$] was one of the major objectives for our methodology because $C_p M$ is the least accurate parameter in the analysis.

While it is also possible to write equation (B.23) in the form (B.24) to give $(k_R'o)_{362}$, this method of analysis is not useful as the range of the extrapolation required is too long ⁽²⁾ [see also ⁽¹³⁾]. For this reason it was recommended in the ICARUS-1 Handbook ⁽²⁾ that $(k_R')_{32}$ should be evaluated at times close to $t = t_2$ using values of $C_p M$ determined from applications of equation (B.21). However, in view of the errors in the determination of $C_p M$, these values of $(k_R')_{32}$ are inevitably less accurate than those of $(k_R')_{22}$ or $(k_R'o)_{262}$ [see also Section A.4].

We note also that care must be taken in carrying out the required linear regression fitting procedures as is illustrated in Section A.4 [see references 4, 5 and 13].

We should observe, furthermore, that equation (B.24) is soundly based [in a mathematical sense] in that the extrapolation to $[\Delta\theta(t) - \Delta\theta(t_2)] = 0$ gives the value of $(k_R'o)_{262}$ at a well defined time, $t = t_2$. This is equally true of all of the coefficients based on "forward" or "backward" integration; however, the starting points for these integrations will usually be chosen to be $t = 0$, $t = t_1$ or $t = T$, and the definition of the heat transfer coefficients at these points is not generally useful. The exception here is the "lower bound heat transfer coefficient, $(k_R'o)_{261}$ " which is also defined at time $t = t_2$. We observe also that $(k_R'o)_{261}$ and $(k_R'o)_{262}$ are the most precise and accurate values of the "lower bound" and "true heat transfer coefficients" which can be derived with the methodology as presently developed. Furthermore,

$$(k_R'o)_{261} = (k_R')_{11} \text{ at } t = t_2 \text{ and } (k_R'o)_{262} = (k_R')_{12} \text{ at } t = t_2 \quad (B.25)$$

so that the best value of $\Delta(k_R')_t$ that can be obtained for use in equation (B.17) is

$$\Delta(k_R')_t = (k_R')_{12} - (k_R')_{11} = (k_R'o)_{262} - (k_R'o)_{261} \quad (B.25A)$$

This "sound basis" of the heat transfer coefficients derived by "forward" and "backward integration" should be contrasted with the corresponding position for the "differential heat transfer coefficients" which has been discussed above.

In the final part of this Section, we need to consider somewhat further the time-dependence of the various forms of the heat transfer coefficient [compare ^{(4),(5),(13),(14)}]. We observe first of all that we are interpreting here the systematic variations of typically 0.4% of the differential or 0.2% of the integral coefficients. The only reason why we are able to investigate systematic variations of such small quantities is the very high precision of the methods of data evaluation.

We observe secondly, that as the differential coefficients are evaluated at local times, they will show the weak time dependence:

$$(k_R') = (k_R'0) [1 - \gamma t] \quad (B.26)$$

[c.f. equations (B.2) and (B.3)]. In the definition of the integral heat transfer coefficients given in this Section, (k_R') has been regarded as being constant whereas the investigation of the differential heat transfer coefficients shows that we should really include the time-dependence, equation (B.26), i.e. we must use equation (B.2) in the integrations. Integration of this equation gives

$$(k_R'0) [\int f_1(\theta) d\tau - \gamma \int f_1(\theta) d\tau + \gamma \int \int f_1(\theta) d\tau d\tau] \quad (B.27)$$

If we now regard $f_1(\theta)$ as being constant throughout the measurement cycle [which is a rough approximation for the case of the "lower bound heat transfer coefficients"] then the integral becomes

$$(k_R'0) f_1(\theta) t [1 - \gamma t/2] \quad (B.28)$$

It follows that the heat transfer coefficients given by equations (B.18) and (B.19) are given by

$$(k_R')_{21} = (k_R'0)_{21} [1 + \gamma(T - t)/2] \quad (B.29)$$

and

$$(k_R')_{31} = (k_R'0)_{31} [1 - \gamma t/2] \quad (B.30)$$

within the limits of the approximation. $(k_R'0)_{21}$ and $(k_R'0)_{31}$ are respectively the values of $(k_R')_{21}$ at $t = T$ and of $(k_R')_{31}$ at $t = 0$. It follows that the slopes of the plots of $(k_R')_{21}$ and $(k_R')_{31}$ versus time are roughly one half of the plot of $(k_R')_{11}$ versus time.

Equation (B.27) also shows the way in which we can test whether the characteristics of the Dewar cells can be described by a single, time-independent heat transfer coefficient. Thus evaluation of $(k_R')_{21}$ according to equation (B.18) gives us the heat transfer coefficient

$$(k_R')_{21} = (k_R'0)_{21} [1 - \gamma T + \gamma \int_T^t \int_T^t f_1(\theta) d\tau d\tau / \int_T^t f_1(\theta) d\tau] \quad (B.31)$$

so that the "time-independent heat transfer coefficient, $(k_R'o)_{21}$ " is readily determined. The fact that heat transfer from the cells can be represented by such a single time-independent heat transfer coefficient has been demonstrated several times [e.g. see Fig. 51 of reference 13]. Indeed, such representations are the basis of our statement that the "integral lower bound heat transfer coefficients" can be determined with a precision given by relative errors of less than 0.01% [see Footnote C.22].

The variations of $(k_R')_{11}$, $(k_R')_{21}$ and $(k_R')_{31}$ with time show that this time dependence of the heat transfer coefficients must be taken into account in evaluations of the rates of excess enthalpy generation aiming at the highest achievable accuracy. If this is not done, then the values of the heat transfer coefficients at the mid-points $t = t_2$, should be used. In that case the values of the rates of excess enthalpy generation calculated will be slightly too small for $t < t_2$ and slightly too large for $t > t_2$. However, the total excess enthalpy calculated for a complete measurement cycle will be approximately correct [see Footnote C.23]. We must also note that the differential heat transfer coefficient, $(k_R')_{12}$, must be used in the evaluations of the rates of excess enthalpy generation and the integral heat transfer coefficients in the evaluation of the excess enthalpy [including the total excess enthalpy for complete measurement cycles]. In particular, the use of $(k_R')_{22}$ in the evaluation of the rates of excess enthalpy generation, will underestimate these quantities [see Footnote C.24].

B.4 The Specification of the ICARUS-1 Data Evaluation Procedures and Experimental Protocols

The modeling of the ICARUS-1 Type Calorimeters, Fig. A.1 has been investigated repeatedly by means of the evaluation of data sets for appropriate "blank experiments" [using in the main Pt-cathodes polarized in D_2O -based electrolytes]. The objective here has been the definition of the appropriate "instrument function". It has been found that this can be accurately defined by equation (B.1).

The next step in this initial phase of the work has been the definition of a set of heat transfer coefficients, which characterize the behavior of the calorimeters and the investigation of their precision and accuracy leading up to their use in evaluating the "raw data sets" of the experimental measurement cycles. The "raw data" used in these investigations have been both those for the appropriate "blank experiments" and those generated by simulations of the cell behavior. An illustration of this phase of the investigation has been given ⁽¹³⁾ [see also references 4, 5, 9, 10, and 14].

The outcome of these investigations has been the demonstration that it is useful to determine first of all the time-dependence of the "differential lower bound heat transfer coefficient, $(k_R')_{11}$ ", as well as of the derived means, $(\overline{k_R'})_{11}$ and $(\overline{\overline{k_R'}})_{11}$. However, these coefficients have a limited precision because their evaluation requires the differentiation of the inherently "noisy" experimental data. Precise and accurate evaluations are therefore best based on the "integral lower bound heat transfer coefficient, $(k_R')_{21}$ " and the "integral true heat transfer coefficient, $(k_R')_{22}$ " as well as on the values $(k_R'o)_{251}$ and $(k_R'o)_{252}$ derived in the extrapolation procedures. These extrapolation

procedures lead both to the elimination of the effects of the water equivalent, $C_p M$, on their values as well as to reasonably accurate determinations of $C_p M$. The differences between the "true" and "lower bound heat transfer coefficients", $(k_R')_{22} - (k_R')_{11}$ or $(k_R'o)_{252} - (k_R'o)_{251}$ can then be used to define the "differential true heat transfer coefficient, $(k_R')_{12}$ ".

It has been found that the precision and accuracy of the integral heat transfer coefficients is so high, that it is possible to investigate their systematic variations with time [typically the systematic variations of just $\sim 0.2\%$ of their numerical values]. Furthermore, it is possible to reduce such data to a single, time independent heat transfer coefficient, e.g. of $(k_R'o)_{21}$ with relative errors below 0.01% . This result is hardly surprising: the "physics" of the calorimeters are quite simple [they are "ideal well-stirred tanks"] and the errors are mainly those set by the temperature measurements. It is also relatively straightforward to specify the changes, which would need to be made to reduce the errors further - say to 0.001% - if that should ever prove to be necessary or desirable.

Although the precision and accuracy of the heat transfer coefficients based on the "forward" integration procedures, $(k_R')_{31}$ and $(k_R')_{32}$, was known to be lower than those based on the "backward" integrations, $(k_R')_{21}$ and $(k_R')_{22}$, the ICARUS-1 Methodology was nevertheless based on such "forward" integrations ⁽²⁾ because such "forward" integration was easier to implement and to combine with the evaluations of the data sets. It was anticipated that the extension of the measurement cycles from 1 to 2 days and, in particular of the calibration periods from 6 to 12 hours, would allow the determination of $(k_R')_{31}$ and $(k_R')_{32}$ with the required and specified precisions and accuracies ⁽²⁾. These changes in the measurement cycles were also expected to facilitate other parts of the investigation such as the determination of the "true heat transfer coefficient $(k_R')_2$ ". The production of plots of the "raw data" and the inspection of these plots leading to the graphical evaluation of $(k_R')_1$ and $(k_R')_2$ were to be the first step in the data processing.

Unfortunately, the protocols laid down in the Handbook for the ICARUS-1 System ⁽²⁾ were not followed in the experiments carried out by the team at NHE [see Footnote C.25]. Furthermore, following the receipt of the first set of data for experiments carried out in the Sapporo Laboratories, it became clear that there were timing errors in the ICARUS-1 System ⁽³⁾. These timing errors did not affect the determination of $(k_R')_{21}$ and $(k_R')_{22}$ [see Footnote C.26]. It was therefore recommended ⁽³⁾ that the protocol set down in the Handbook ⁽²⁾ be strictly adhered to, that the preliminary evaluations should be based on $(k_R')_1$, $(k_R')_2$ and $(k_R')_{11}$ and that the final evaluation should be based on $(k_R')_{21}$, $(k_R')_{22}$, $(k_R'o)_{251}$ and $(k_R'o)_{252}$. It is evident that these instructions were ignored.

The development of the various aspects of the data analysis described in the sub-Section B.4 is illustrated in part by the analysis of the experiment described in Section A.4. Inevitably, this illustration is incomplete because of the very early development of "positive feedback" in this experiment.

Section C

As is explained in the main text, Section A, Section C contains a set of Footnotes and Comments which should be read in association with the main text.

- C.1. It is important to understand that the ICARUS 1 System ⁽²⁾ [completed in December 1993] used a Keithley 199 scanning DMM unit and that this system introduced timing shifts into the data acquisition processes [the well-known multiuser-multitasking problem associated with the use of PC's]. This applies equally to all other experiments carried out before ~ 1st January 1995. These timing errors did not affect evaluations based on $(k_R')_1$, $(k_R')_2$, $(k_R')_{11}$ or $(k_R')_{12}$ but had to be taken into account in some evaluations at higher levels of precision and accuracy based on $(k_R')_{21}$ and $(k_R')_{22}$; they were especially important in the evaluations of $(k_R')_{31}$ and $(k_R')_{32}$ as was explained especially in three reports ^{(3),(4),(5)} [for the definition and use of these heat transfer coefficients, see Sections A and B].

There is also extensive further correspondence dealing with the analyses of Data Sets collected with the ICARUS-1 and 2 Systems. However, the three reports ^{(3),(4),(5)} are sufficient to explain the reasons for the choices of the methodologies.

- C.2. The two Reports ^{(3),(4)} were sent in the first place to Technova [see also Footnote D.2].
- C.3. It should be noted that it is possible to achieve accurate control of the temperature of the heat sink by using this simple design. In turn this allowed the installation of a large number of experiments at low cost.

There are misleading statements in the literature about this aspect of the experiment design. In fact, the design follows the common strategy of using two thermal impedances in series, a strategy, which is required for experiments aiming at high accuracy, [however, see also Section A.2].

- C.4. The major reason is that this is the only experiment to date where we have a listing of the raw data together with the evaluation carried out according to the procedures used in the NHE Laboratory. This is evidently of key importance to (i) and (ii) especially in view of the assessment of the ICARUS methodology which has been published ⁽¹¹⁾ [see also Footnote D.3].

- C.5. (via) several "blank experiments" were to be carried out [at least one for each cell in use; the use of Pt-cathodes in 0.1M LiOD/D₂O was recommended]. The experiments were to be carried out at a single current density using otherwise the protocol described in (ia)-(va);

(viia) the execution of the "blank experiments" was to be followed by experiments using cathodes made of Johnson Matthey Material, Type A.

The protocols (ia) - (va) were again to be followed;

- (viiiia) the first step in the data evaluation was to be the plotting of A3 or A4 sized graphs of the "raw data". The heat transfer coefficients $(k_R')_1$ and $(k_R')_2$ were to be derived for each measurement cycle [see Section B].
- (ixa) the next step was to be the construction of $(k_R')_{11}$ - type spreadsheets coupled to the determination and interpretation of $(k_R')_{11}$, $(\overline{k_R'})_{11}$ and $(\overline{\overline{k_R'}})_{11}$ [see Section B.3]. The further evaluation of these spreadsheets was not specified in 1993; this was a matter which was to be decided by a collaborative program between NHE and I.M.R.A. Europe.
- (xa) after the execution of (viiiia) and (ixa) the $(k_R')_{21}$ - type spreadsheets were to be prepared and values of $(k_R'o)_{261}$ and $(k_R'o)_{361}$ and the associated values of C_pM were to be determined. These values of C_pM were to be used both to correct the evaluations in (ixa) and to determine the "true heat transfer coefficients, $(k_R')_{32}$ " at times close to the end of the calibration period. Following the receipt of the first set of "raw data" collected with the ICARUS-1 system ⁽³⁾ this objective was changed to the evaluation of $(k_R')_{22}$ and of $(k_R'o)_{262}$ at times close to the end of the calibration period.
- (xia) it was envisaged that, following the completion of this initial stage of the investigation, the ICARUS program would move on to the examination of materials variables as well as the production of ICARUS-2. This second part was intended to deal also with the effects of increasing the current density, the analysis of data close to the boiling point [including boiling episodes] and "Heat-after-Death". The effects of changes of current density were to be followed in line with earlier investigations ^{(15),(16)}, see (iiia), using an initial polarization at low current densities for various periods of time followed by a raising of the current density sufficient to drive the systems through the region of "positive feedback" and, eventually, towards the boiling point. The analysis of data close to the boiling point and of "Heat-after-Death" was to be covered initially by appropriate "Case Studies". The material given in Section A7 - 9 goes some way towards fulfilling the needs of such "Case Studies".

The topic of "positive feedback" was not included in the ICARUS-1 program mainly because these effects greatly complicate the analyses [see Section A.3 and A.4] but also because aspects of this topic were still being actively investigated at IMRA Europe [see Footnote E.11]. However, extensive comments on this topic were included in the Report ⁽⁴⁾ on the second set of experiments carried out by NHE as the systems had evidently been driven into the regions of "positive feedback" [see also ⁽⁸⁾].

C.6. However, the material outlined in ⁽¹³⁾ was also contained in earlier letters and reports sent to Japan and, in part, in papers, which have been published previously.

C.7. Reference 13 deals first of all with the analysis of a data set generated by simulation using the most the drastic simplifications of the differential equation representing these Fleischmann-Pons calorimeters. The second part deals with the analysis of a blank experiment [Pt cathode] polarized in 0.1 M LiOD/D₂O. This full report in as received condition [from Fleischmann] consists of 190 pages and will be published in another report (see Ref. 13).

C.8. We note here that the values of the rates of evaporative loss given in the spreadsheets contained in the Report have been calculated with an earlier version of equation (B.4) using the values

$$\begin{array}{rcl} L & = & 41,673.7 \text{ J Mole}^{-1} \\ F & = & 96,484 \text{ C Mole}^{-1} \end{array}$$

At temperatures close to the boiling point the values of P^* and θ_{boiling} were adjusted as described in Section B.2.

C.9. Changes in the parameter listing are much more important for other parts of the evaluation e.g. the adjustment of the heavy water equivalent, $C_p M$ of the cells in the evaluation of the lower bound heat transfer coefficients, $(k_R)_{11}$ [see Section A.3].

C.10. Independent calibrations show that the heat transfer coefficient for cells filled with air are about 0.75 of the values of these coefficients for cells filled with liquid ^{(9),(10)}. It follows therefore that the marked increase in the heat capacitance of the cells filled with D₂O vapor at temperatures close to the boiling point must lead to the maintenance of the heat transfer coefficient at the value which applies to cells filled with liquid.

C.11. The group at the NHE Laboratory attempted to determine the values of ΔM directly by adding a condensation section to the cells [see Footnote D.5].

C.12. As has been explained previously ⁽¹³⁾, we had hoped to circumvent the need for such an extended description so as to avoid overburdening our accounts with redundant symbolism. In fact, the Handbooks ^{(1),(2)} and earlier Reports ^{(3),(4),(5)} used a simplified description. Again, as has been described previously ⁽¹³⁾, it is the deviation by the NHE group from the recommended protocols and procedures and their use of inaccurate and/or invalid methods of data evaluation ⁽¹¹⁾ which makes it necessary to use the more closely defined description, (B.8).

C.13. Of course, other averages can be made but the use of the 11-point average has been found to be especially useful.

C.14. The value of $C_p M = 490 \text{ JK}^{-1}$ was appropriate for somewhat larger cells used in an earlier phase of our investigations.

- C.15. Objections have often been raised to the procedures which we have adopted based on the fact that it is alleged that we have not "binned the data" i.e. we have not signal averaged before the data analysis [see Footnote D.7]. However, "binning of the data" must always be approached with great caution: one should only "bin data" or "bin coefficients" if these data or coefficients can be expected to be constant over the averaging interval. This is not the case for $(k_R')_{11}$ unless the effects of the term $C_p M(d\Delta\theta/dt)$ have been taken into account. Once this is done we can, of course, bin the coefficients as we have done in deriving $(\overline{k_R'})_{11}$ and $(\overline{k_R'})_{11}$.

In the case of the interpretation of data obtained with calorimeters relying on radiative cooling, the position is further complicated by the fact that the differential equation representing the calorimeters, equation (B.1), is both non-linear and inhomogeneous. It does not follow therefore that the coefficients derived by averaging the data will be the same as the averages of the coefficients derived by using the "raw data". However, it was confirmed in 1992 [when the approach outlined here was initiated] that one does, in fact, obtain an equivalence provided attention is restricted to regions where $(d\Delta\theta/dt)$ is adequately small. It was concluded that in such regions the differential equation (B.1) could be sufficiently linearised and that the second order small differences were sufficiently small to allow such averaging. However, as the values of $(k_R')_{11}$ obtained in this way were identical to those obtained using the procedure outlined in Section B.3, there was evidently no justification in pursuing the matter further [nor to complicate the instrumentation!].

It is evident therefore that these particular criticisms were invalid [see Footnote E.4].

Averaging procedures are considered further in Section B.3 in the discussion of heat transfer coefficients based on the forward and backward integration procedures.

- C.16. A similar comment applies to the determination of $(k_R'o)_{161}$; the time at which $(d\Delta\theta/dt) = 0$ will usually be accessible for experiments in which $t_1 = 9$ hours. However, no such point can be defined for $(k_R'o)_{171}$ so that this determination is mathematically questionable; this is therefore equally true for $(k_R'o)_{181}$ although these extrapolations are certainly sound from an operational point of view.
- C.17. The validity of equation (B.17) was established at the time of construction of the ICARUS-2 System.
- C.18. In our early work [prior to October 1989] we had overcome this particular difficulty by using $(k_R')_2$ as starting values for the non-linear regression procedure leading to the "true heat transfer coefficient, $(k_R')_5$ ". Here we fitted the numerical integrals of the differential equation (B.1) to the data sets for complete

measurement cycles. The relative errors in $(k_R')_5$ which we could achieve in this way were below 0.1% even when using the unsatisfactory early version of our calorimeters [i.e. those not silvered in their top portions].

This use of the non-linear regression procedure had the further distinct advantage that we could adjust the integration interval in regions where the temperature was varying rapidly with time so as to achieve the required accuracy in the integrals. This was not possible for the methods leading to $(k_R')_{21}$, $(k_R')_{22}$, $(k_R')_{31}$ and $(k_R')_{32}$ discussed in Section B.3 when using the ICARUS-1 and -2 Systems because the intervals for data acquisition were fixed. Indeed, the interval 300 s was chosen because such an interval does not degrade the evaluation of any of the series $(k_R')_{21}$, $(k_R')_{22}$, $(k_R'o)_{251}$, $(k_R'o)_{252}$, $(k_R'o)_{261}$, $(k_R'o)_{262}$, $(k_R'o)_{271}$ and $(k_R'o)_{272}$. However, it does degrade the evaluation of $(k_R')_{31}$, $(k_R'o)_{351}$, $(k_R'o)_{361}$ and $(k_R'o)_{371}$ to some extent and leads to a marked degradation of $(k_R')_{32}$, $(k_R'o)_{352}$, $(k_R'o)_{362}$, and $(k_R'o)_{372}$. The fact that the data acquisition interval was too long for straightforward estimates of the $(k_R')_{i,j,1}$ series of heat transfer coefficients was already pointed out to NHE in the ICARUS-1 Handbook ⁽²⁾ [see Footnote E.6].

We have pointed out elsewhere ^{(15),(16)} that a major reason for our choice of non-linear regression was that the pressure of events in 1989 did not allow us to go through the logical sequence of using linear regression, multi-linear regression and non-linear regression [in fact, we had to opt for a "catch-all" methodology and any integrated version of equation (B.1) was certainly expected to require the use of non-linear regression]. However, our use of this methodology was certainly not understood ^{(18),(19)} [see Footnote E.7] and we also could not make non-linear regression "user friendly" with the computing power available to us at that time. In 1991-92 we therefore investigated the application of linear regression ^{(15),(16)} and this became part and parcel of the ICARUS-1 methodology ⁽²⁾.

- C.19. It is in any event necessary to change the methodology of the evaluation in view of the early onset of "positive feedback" see Section A.4.
- C.20. As far as one is able to tell from the limited amount of information available.
- C.21. We retained the designation 22 as a flag to indicate that the "backward integration" methodology was the primary objective for accurate evaluations.
- C.22. Reference ⁽¹³⁾ contains extensive discussions of the errors of the various heat transfer coefficients and the causes of these errors. This reference will be published in another report.
- C.23. This will explain both our strategies for determining the heat transfer coefficients [which give the values at $t = t_2$] as well as giving a further reason for choosing 2-day measurement cycles with t_2 corresponding to the end of Day 1.
- C.24. The need for posing this restriction was pointed out repeatedly. However, we

note that the group at NHE used integral heat transfer coefficients to evaluate the rates of excess enthalpy generation⁽¹¹⁾ and this applies equally to their spreadsheets for the experiment considered in this Report.

- C.25. Or, at least, they were not followed systematically.
- C.26. The points at which data were acquired were always correctly timed. This is true for all the experiments, which we have carried out [see Footnote E.8].
- C.27. and as has been pointed out elsewhere, we no longer have the "raw data" for these experiments.
- C.28. It is therefore impossible to carry out the tests based on the application of the protocols specified for these experiments, especially those based on (iia) see Section A.2.
- C.29. However, we should note that as far as the results for this experiment are concerned, the early development of "positive feedback" would in any event have precluded the evaluation of $(k_R')_2$ [see Sections A.3 and 4].
- C.30. We should make a special note here of the fact that the analyses presented in this report are incomplete in that the effects of these changes in current density have not been considered except for that on Day 26 which has been discussed as a special case of "Heat-after-Death", see Section A.10. In fact, it is possible to develop analyses analogous to those leading to $(k_R')_{22}$, $(k_R')_{262}$, $(k_R')_{32}$ and $(k_R')_{362}$ but based on the perturbations due to changes of current density. This is a matter, which should be considered further in an addendum to this Report.

In this connection it is of some interest that the analysis of the effects of changes of current density in one of the data sets of the investigation by the group in Harwell⁽²²⁾ gives a clear demonstration of excess enthalpy generation. A paper dealing with this topic⁽²³⁾ was rejected for publication [two further papers dealing with the reanalysis of data sets generated by this investigation have also been rejected for publication].

- C.31. A phenomenon which has also been observed and commented on by other authors.
- C.32. It has been argued^{(6),(7)} that this phenomenon is linked to the need to achieve high levels of loading of the cathodes by D^+ which is probably also associated with a change from exo- to endothermic absorption; an alternative explanation⁽²⁴⁾ is that these phenomena are linked to the formation of a new phase, the γ -phase⁽²⁵⁾.

It is not clear at the present time whether [and if so how] the onset of "positive feedback" is linked to the increase of cell temperature and the results of the experiment certainly argue against such an interpretation [see Sections A.3 and

A.4]. However, it seems to be reasonably clear that increases of temperature facilitate the phenomenon. Furthermore, maintenance of the electrode in the region of the onset of "positive feedback" [current density, temperature] leads to the onset of oscillatory phenomena ^{(6),(7),(24)} which reduce and may even eliminate the effect. It is the sensitivity of "positive feedback" to the operating conditions, which is a major factor in making the outcome of experiments [i.e. the level of the rates of excess enthalpy generation] dependent on the experimental protocols.

- C.33. We have to take note of the fact that the usable range of current densities was necessarily restricted by the maximum output of the galvanostat [± 1 A at ± 100 V] and the relatively large surface area of the electrode [3.15 cm^2].
- C.34. Furthermore, the current densities remained in the low range for the observation of excess enthalpy generation ⁽¹²⁾.
- C.35. A correction to Fig. A.4 may form an addendum to this Report.
- C.36. The effects of such changes may form a further addendum to this Report.
- C.37. Which furthermore quoted results showing that the volumes of the electrolyzed gases agreed with those calculated for electrolysis according to Faraday's Laws.
- C.38. Nevertheless, the recombination of D_2 and O_2 formed in the electrolysis continues to be invoked in explanations of the excess enthalpy generation observed e.g. ⁽¹¹⁾ and the numerous comments by *inter alia* D. Morrison and S. Jones.
- C.39. The conditions of operation above Day 62 need to be investigated further.
- C.40. Hypothetical differences in temperature due to inadequate mixing have frequently been invoked in hypothetical arguments about the performance of these calorimeters. We note, however, that inadequate mixing would not give rise to a systematic and time invariant difference in temperature between the two positions. Moreover, such differences in temperature would not be expected because the thermal relaxation time, $\tau = C_p M / 4(k_R') \theta_{\text{cell}}^3$, is of order 5000 s whereas the radial and axial mixing times are ~ 3 and ~ 20 s [as revealed by tracer experiments]. Small differences in the cell temperature can only be observed in the vicinity of the electrodes and calibration resistor i.e. within the Prandtl boundary layers. However, the volumes of liquid within these boundary layers are negligibly small compared to the volume of electrolyte in the cells.
- C.41. In considering the precision and accuracy of the measurements of the "raw data" we should note that the cell currents, column 6 of Table A.2, have been rounded to 5 significant figures. These currents should, in fact, have been given to 6 significant figures.
- C.42. With a thermal relaxation time of 5000 s, the temperature perturbation will only

have reached 98.67% of its final value within the calibration period. The consequent 1.33% error is outside the limits specified for the ICARUS Systems [see also Section A.4].

- C.43. Such effects can be seen in the "raw data" of some of the experiments carried out by the group at Harwell ⁽²²⁾ as well as in some of the early measurements carried out by the group at NHE ^{(4),(5),(24)}.
- C.44. This is a feature, which will be common to all calorimetric systems used to investigate a thermal source subject to "positive feedback". [Moreover it is a feature of any system subject to "positive feedback"]. It is likely that the neglect of this fact is responsible for much of the confusion in the research on "Cold Fusion".
- C.45. The spreadsheets contain the following columns of entries:
- 1) the time of the data acquisition, t/s;
 - 2) $\theta_{\text{cell, short thermistor}}$, the temperature measured with the "short thermistor"/K, see Fig. A.1;
 - 3) $\theta_{\text{cell, long thermistor}}$, the temperature measured with the "long thermistor"/K, see Fig. A.1;
 - 4) E_{cell}/V ;
 - 5) I_{cell}/A ;
 - 6) θ_{bath} , the temperature of the water bath, /K;
 - 7) $(d\theta_{\text{cell}}/dt)_{\text{short thermistor}}$, /K s⁻¹;
 - 8) the enthalpy input, /W;
 - 9) the rate of evaporative cooling, /W;
 - 10) the Joule heat input due to the resistive heater, /W;
 - 11) $f_1(\theta)_{\text{short thermistor}}/\text{K}^4$;
 - 12) the rate of excess enthalpy generation, /W;
 - 13) $10^9 (k_R')_{11}$, /WK⁻⁴;
 - 14) an entry related to the water equivalent of the calorimeter, /JK⁻¹;
 - 15) $(d\theta/dt)_{\text{long thermistor}}$, /K s⁻¹;
 - 16) the rate of evaporative cooling, /W;
 - 17) $f_1(\theta)_{\text{long thermistor}}/\text{K}^4$;
 - 18) the rate of excess enthalpy generation, /W;
 - 19) $10^9 (k_R')_{11}$ /WK⁻⁴;
 - 20) an entry related to the water equivalent of the calorimeter, /JK⁻¹.

It is evident that the derived data in columns (7), (9), (11), (12) and (13) are based on the temperature measured with the "short thermistor", column (2), while those in columns (15), (16), (17), (18) and (19) are based on that measured with the "long thermistor", column 3. The basis of the entries in columns (14) and (20) is not clear, nor is it apparent how this information may have been used.

- C.46. The water thermostats surrounding the cells were run at ~ 30°C in the early work

in Salt Lake City. In 1988 attempts were made to allow for this shift in the reference temperature as well as the fact that electrolysis takes place from 0.1 M LiOD in D₂O and not D₂O itself. While the thermoneutral potential is certainly not 1.527 V, it is closer to this value than to 1.54 V. The value 1.527 V has therefore been used in the calculations summarized in Tables A.3 - A.9.

- C.47. It appears that the facilities for monitoring the current to the calibration resistor and the voltage across this resistor were disabled.
- C.48. More exactly, whether the values of $(k_R')_{11}$ plotted versus time fall on a common straight line as shown in e.g. ^{(4),(13),(14)}, see also Fig. A.20 in Section A.5.
- C.49. In actual fact, the matter is more complicated than has been indicated in the main text. In the method originally proposed by NHE the "lower bound heat transfer coefficient" determined before the application of the calibration pulse was used in attempts to derive the rate of excess enthalpy generation during the application of this pulse. It is not surprising that such a method can only give the correct result provided there is a zero rate of excess enthalpy generation for the period $t < t_1$ before the application of the pulse [as well as for $t > t_2$ following the termination of the pulse] while the calibration pulse itself [during $t_1 < t < t_2$] leads to the generation of excess enthalpy. These facts were illustrated using simulated data ⁽⁴⁾ but the Report did not lead to any further discussion.

It is of interest that the behavior of this Pd-B study during Day 3 to some extent satisfies the preconditions for the application of the NHE methodology [see the main text]. It is also of interest that the method proposed by NHE bears some resemblance to an evaluation used previously ^{(15),(16)} except that the earlier method used the maximum value of the "lower bound heat transfer coefficient, $(k_R')_{11}$ " observed during the early stages of the experiment. The use of such a value of the "lower bound heat transfer coefficient" is discussed further in Section A.6.

- C.50. It so happens that there is some validity to this conclusion due to the influence of "positive feedback". This matter is discussed in Section A.4.
- C.51. It is evidently very important to establish whether this early onset of "positive feedback" is a property of the Pd-B alloy used in this experiment. It should be noted that it was proposed some years ago that Pd-Ag alloys used in diffusion tube separators should be replaced by Pd-B alloys. Evidently Pd-B alloys were expected to display some beneficial properties, and the literature on this topic should be searched and the main manufacturers should be consulted.
- C.52. The steps outlined in Section A.4 do not correspond to the sequence in the ICARUS-style evaluation ⁽²⁾. In that evaluation [as modified in ⁽³⁾] the first step is the preparation and inspection of A-4 sized plots of the "raw data" leading to the evaluation of $(k_R')_1$ and $(k_R')_2$; the second step is the preparation and interpretation of $(k_R')_{11}$ - spreadsheets while the third step is the preparation and

evaluation of $(k_R')_{21}/(k_R')_{31}$ - spreadsheets leading up to the evaluation of $(k_R'o)_{261}$, $(k_R')_{262}$, $(k_R')_{361}$ and $(k_R')_{362}$. It should also be noted that the ICARUS software prepares the more restricted version of the $(k_R')_{21}/(k_R')_{31}$ - spreadsheets [restricted to $t_1 < t < t_2$] as shown in Table A.5 and not the version for the whole measurement cycle $0 < t < T$ shown in Table A.4.

It should be noted furthermore that the tables presented in this Report only contain a part of the information of the ICARUS-style spreadsheets.

- C.53. As has been noted in Section B.3, it is possible that this insertion of additional data points has not been carried out in the systems supplied to NHE. This particular question could only be resolved by re-implementing the relevant software.
- C.54. The values for the first 20 - 30 points must be excluded as the benefits of using the integral coefficients are only established with increasing time. Similarly, the first 20 - 30 points must be excluded if the interpretation is based on backward integration i.e. if we consider $(k_R')_{21}$.
- C.55. More exactly the evaluation of $(k_R')_{32}$ was restricted to times close to t_2 for calibration periods of 12 hours i.e. for measurement cycles lasting 2 days ⁽²⁾.
- C.56. The increase of the pseudoradiative heat transfer coefficient due to the "over filling" of the cells is important from a different point of view. It has been shown in Section B.2 that the thermal capacity of the head space in the cells [filled with the mixture of deuterium, oxygen and D₂O vapor] must increase markedly above that predicted for the head space filled with deuterium and oxygen alone at temperatures close to the boiling point of the electrolyte. It follows, therefore, that heat transfer from the cell must be controlled by the coefficient for cells "overfilled" with electrolyte. This matter is considered further in Section A.6.
- C.57. The main question is that if the charging of the electrode is indeed 100% efficient and, if this charging is completed at $\sim 130,500$ s, then the total charge taken up by the electrode is in excess by a factor of five of that predicted for a charging ratio $D/Pd = 1$. Related to this question is the problem that the total excess enthalpy during the charging period [~ 3800 J] is in excess by a factor of two of that which would be predicted for $D/Pd = 1$ and an heat of absorption of 40 kJ mole^{-1} . This suggests anomalous excess enthalpy production during the charging period. Experimentally, vigorous gas evolution was observed after $13,200$ s [$I=0.150\text{A}$]. This corresponds to a loading of $D/Pd=0.5$ at that time.
- C.58. Evidently, heat transfer to the walls of the Dewar cell is maintained by the vapor phase at the very least if this phase is filled with D₂O vapor at temperatures close to the boiling point of the electrolyte [c.f. the increase in the thermal capacity of the vapor phase, Section B.2]. It follows that the heat transfer coefficient for the cell filled with vapor is the same as the heat transfer coefficient for the cell filled

with electrolyte. In actual fact, the heat transfer coefficient for the cell filled with vapor will be that for a cell "overfilled" with electrolyte i.e. some 5% above the value $0.85065 \times 10^{-9} \text{ WK}^{-4}$ [c.f. the discussion of Fig. A.20].

- C.59. It is important to realize that the calculation of the total enthalpies outlined in the main text, Section A.7, is independent of the rate of reflux.
- C.60. It is also important to realize that the interpretation given by NHE shows the other features of excess enthalpy generation to which we have frequently drawn attention e.g. the fall of the "lower bound heat transfer coefficient, $(k_R')_{II}$ ", with time due to the build up of excess enthalpy production. In fact excess enthalpy generation was observed throughout the time range except for the last four data points where the calculation of the rates of evaporative cooling are invalid. These matters passed without comment.
- C.61. The group at NHE attempted to derive the rate of evaporative cooling directly by collecting and weighing the distillate. However, apart from the inaccuracies introduced by the unknown hold-up in the distillation system, the data derived cannot be interpreted in the absence of information about the rate of reflux. It is therefore in any event necessary to carry out the calculation outlined in Section A.7 so that the weighing system does not produce any additional useful information.
- C.62. The operation of the experiment on Day 68 should be contrasted with earlier measurements ^{(15),(16)}. The rates of excess enthalpy generation in these earlier experiments reached much higher values than those reported here [up to two orders higher]. Some of the complications in the evaluation of this experiment could therefore be avoided. In particular, it could be assumed that the last part of the earlier experiments were controlled by a process akin to boiling. This was sufficiently intense that the effects of reflux could be neglected.
- C.63. As has been noted above the calibrations used in an earlier investigation were derived by using a heater spiral spanning the whole volume of the cell i.e. heat was applied uniformly throughout this volume.
- C.64. However, if we describe this as the "conventional wisdom" of the research in this field, then we can see that the results for this experiment fly in the face of this "conventional wisdom". In particular, excess enthalpy generation was observed on Day 3 of the measurement cycles at a current density below the threshold value while "positive feedback" was established at a temperature below this further threshold. We can therefore only regard the criteria used to search for category #1 of the phenomenon of "Heat after Death" as rather "broad brush indicators".
- C.65. Possibly because of the cracking of the electrodes due to the repeated loading and de-loading.

- C.66. The non-uniform charging of electrodes is probably responsible for the marked increase in the rate of excess enthalpy generation at temperatures approaching the boiling point. The electrodes then become covered by a porous film and charging takes place at very high current densities through the pores in the film. This process is favored by the "tertiary current distribution" in the film: the very high current densities lead to increases in the OD^- concentration in the pores which in turn enhances the non-uniform current distribution.

A program of work on the non-uniform charging of electrodes [using *inter alia* electrodes partially covered by diffusion barriers] could not be established at IMRA Europe because of lack of resources. This is probably one of the most important neglected aspects of the work because, if such charging should prove to be successful, it would allow the construction of energy efficient systems.

- C.67. It is unlikely that the variation of the distillate with time [as determined in the NHE investigations] could be usefully interpreted.

Section D

As is explained in the main text, Section A, Section D contains a set of Footnotes and Comments, which should not necessarily be included in a Report given extensive circulation.

- D.1. [see also Footnote C.1] As has been explained in earlier correspondence, M. Fleischmann only obtained a copy of the Handbook describing the ICARUS-2 System ⁽¹⁾ indirectly during 1998 from M.H. Miles. Prior to this, M. Fleischmann was denied access to this Document.
- D.2. [see also Footnote C.2] It is not clear whether NHE ever took any account of these reports [or of the extensive further correspondence] as their receipt was never acknowledged. Therefore, there was no correspondence concerning their contents. However, we note that the whole project was to be carried out in the Public Domain. Presumably, therefore, these Reports should be made available to "third parties" if this should be required.
- D.3. [see also Footnote C.4] At the 7th International Conference on Cold Fusion, one of us [M. Fleischmann] was also given a CD containing the raw data for a series of experiments carried out in the NHE Laboratories. This CD may also contain information about the evaluations, which were carried out. However, none of these experiments correspond to the ones, which had originally been requested [see ⁽⁵⁾ and the preceding reports and correspondence]. These experiments were all carried out prior to June 1994 [although some of the experiments were continued to later dates]. It had originally been intended to use this date as a "cut-off" point [mainly because we do not believe that there has been any significant advance

since that time, see also Footnote E.1].

- D.4. The possibility of the "overfilling" of the cells was to have been avoided by adding an audio alarm system to the "switching boxes" which were part of the ICARUS-2 System. This modification was never included and as such it was never used.
- D.5. It was difficult to see how anybody could convince themselves that such measurements could give any meaningful results. The amount distilled was determined by weighing the distillate and, in consequence, was reduced by the hold-up in the vapor filled section of the cell as well as the condensation unit. Furthermore, the rate of evaporation had to be derived by differentiating the "noisy" experimental data. However, let us suppose that these defects could have been overcome. In that case we would at best have derived information about the reflux ratio, a quantity which does not give any useful information about the rate of excess enthalpy generation. The only directly useable information is the detection of the time at which the cells are driven to dryness. However, this time can be determined directly from the "raw data" by noting the fall in the cell current or by direct visual observation or, better, by using video recordings [as we had done in 1992 ^{(15),(16)}].

This part of the instrumentation was also constructed at IMRA Europe and the effort involved caused considerable disruption of the research program [see Footnote E.2].

- D.6. Furthermore, it is possible to use filter functions other than the "square-box" filter of the 11-point averaging and to design such filters to facilitate the evaluation/demonstration of specific points of interest [e.g. the effects of "positive feedback"]. A program of work on these aspects was started in 1992 but had to be abandoned because of lack of time and resources [see also Footnote E.3].
- D.7. See especially the comments of Stephen Jones.
- D.8. It is important here to draw attention to a shortcoming of the modeling of the calorimeters, equation (B.1), as well as the evaluation of the heat transfer coefficients [especially of $(k_R)_{11}$ and $(k_R')_{12}$]. In this modeling and in these evaluations we assume that the fluctuations in the temperature about the relevant mean in the time-domain are scaled by the total water equivalent of the calorimeter, $C_p M$, to give the fluctuations in the enthalpy content of the system. It may well be that this is an overestimate and that the temperature fluctuations take place in just a small element of the liquid in the calorimeter.

The detailed consideration of this topic is beyond the scope of the present report. However, we should note that the correct calculation of the fluctuations in the enthalpy content requires both temperature measurements at many locations coupled to the evaluation of proper weighted means and the development of the

theory of the power spectral densities of the fluctuations. These topics were investigated during 1989 but were subsequently abandoned because of lack of resources and because it became evident that $(k_R')_{11}$ and $(k_R')_{12}$ could be derived within the target error limits by taking appropriate averages [e.g. $(\overline{k_R'})_{11}$ or $(\overline{k_R'})_{11}$]; this was equally true of the integral heat transfer coefficients. Furthermore, it was also clear that it would be possible to obtain the required spatial averages by using the ICARUS-14 calorimeters [originally described as the ICARUS-4 versions].

We note here that the work carried out in 1989 relied on the calculation of the autocorrelation function of the fluctuations. However, it became clear that it was necessary to model the cross-correlation functions and that the problem of the generation of thermal noise in calorimeters was formally similar to the problem of noise generation by aerofoils.

Section E

For reasons which will become apparent, this Section contains a set of Footnotes and Comments which perhaps should not be included in the main Report.

- E.1. One of the authors [M. Fleischmann] has explained on other occasions that all the data sets which were originally in his possession were removed from the material returned to him from France. This includes all the important data sets collected in the NHE Laboratory prior to June 1994, details of a very important experiment carried out in another laboratory in Japan which mimics the procedure which had been used in Utah prior to October 1989 [i.e. the data analyzed in ⁽¹²⁾], the "Harwell" data sets and all information about the experiments carried out in the National Cold Fusion Center, Salt Lake City. The only information now in our possession consists of parts of data sets, which had been evaluated at various times during M. Fleischmann's stays in the U.K.

It might well prove possible to reconstruct at least parts of the original data sets from this fragmentary material. However, it is not clear at this time whether such data sets could be evaluated without access to the relevant laboratory notebooks. We note in this connection that one of us [M. Fleischmann] has frequently requested access to this information and has suggested procedures for the further evaluations. These requests have always either been ignored or else blocked.

- E.2. We expressed our reservations repeatedly about this part of the research program but never received a reply. It should be noted that this development took place following completion [in June 1994] of the first part of our investigations of the long-term maintenance of boiling conditions using the calorimeters in the

sequence ICARUS-4 to ICARUS-9 ⁽¹⁷⁾. It was this part of the program, which needed to be developed further, and the ICARUS 10-13 Calorimeters were designed and constructed with this end in view. However, these calorimeters were never put into use.

- E.3. Part of this program was aimed at self-tuning control of the systems with a view to implementing automatic "hill-climbing" to increase the enthalpy output.
- E.4. This particular critique is a minor [but, nevertheless very good] example of much of the criticism in this field. A scenario is set up [or a point is raised] leading to highly critical statements [should one say, vehemently critical statements?]. There is no doubt that these statements would have been justified if the original scenario [or point] had been valid. However, the original scenarios [or points] were not valid and the critique is therefore quite incorrect.

Other good examples of such critiques include statements about the inadequacy of stirring, temperature inhomogeneities in the cells, the definition and use of pseudo conductive or pseudoradiative heat transfer coefficients, the use of a single thermal impedance [as against two thermal impedances in series] the methodologies of evaluating the results etc. etc. All of these initial scenarios should have been checked against the published papers or, perhaps, even been raised in correspondence prior to the decision to publish the critiques?

- E.5. As has been noted previously ⁽¹³⁾, it is important to establish whether the group at NHE ever followed this particular instruction and, if they did so, what conclusions they may have drawn from such plots. However, it is unlikely that the group at NHE ever constructed such plots because, if they had done so, they would surely have understood one reason for the specification in the ICARUS-1 System that the duration of the calibration pulses had to be lengthened to 12 hours [and the reasons for other specifications of durations of the experiment cycles]. They would also have detected the presence [or absence] of "positive feedback". One of the authors [M.H.M.] reports that he saw no evidence for the construction of such plots while he was working at NHE. Inadequate inspection of "raw data" is a malaise, which afflicts much of modern science.
- E.6. However, this did not lead to any discussion of the advantages/short comings of the various methodologies of estimating the "true heat transfer coefficient". We note that the ICARUS-1 System Handbook was described as "Version 1- Low Power Measuring System for Three Cells" with the qualification on page 62: "The experimental equipment and hardware are similar to those which we currently use. It is envisaged that updates of the software will be dependent on the needs of the laboratories taking part in this research program".

We also note that we never received any communication from the group at NHE concerning the methods of data evaluation. It is evident that the group continued to base their evaluations on $(k_R')_{32}$, and it appears that the value of this coefficient

determined from Day 3 was also used for the whole of the evaluation of the experiment MC21 [Pd-B].

E.7. Intentionally?

E.8. This is an aspect of the greatest importance because it shows that it would enable the precise and accurate evaluation of all the data sets, which have been determined hitherto [see Footnote E.9]. In view of the difficulties, which were evidently being experienced in the data evaluation, it has been urged at various times that a group should have been set up charged with the task of evaluating the various data sets. The task of the parent groups would then have been reduced to the validation of these evaluations. There was never any reply to these suggestions [see Footnote E.10].

Short of this suggestion, one of us [M. Fleischmann] urged that the raw data for various selected experiments should be released for further study. However with the exception of this experiment [considered in this Report] and the data sets provided by Dr. Asami at ICCF 7, there has again been no response to these proposals. We note that the data provided at ICCF 7 did not include any of the data sets which had been previously selected. This selection was based on the desire to restrict data evaluations to experiments carried out prior to June 1994 [with the important exception of this experiment].

E.9. It would evidently have been sensible to restrict such evaluations to just a few data sets because further work should have been carried out using ICARUS-14 Calorimeters [originally classified as the ICARUS-4 Systems].

E.10. However, we note that neither the original authors nor Harwell commented on the reassessments ^{(20),(21)} of parts of the original investigation by the group at Harwell ⁽²²⁾.

E.11. Furthermore, one of us [M. Fleischmann] had believed for some time that lines of research based on this work could point the way towards aspects of importance to National Security and should therefore be excluded from work in the Public Domain [see also Footnote E.12]. It should be emphasized here that the topic on its own does not point to matters of interest to National Security.

E.12. The text of this Footnote may be made available on application.

Acknowledgement

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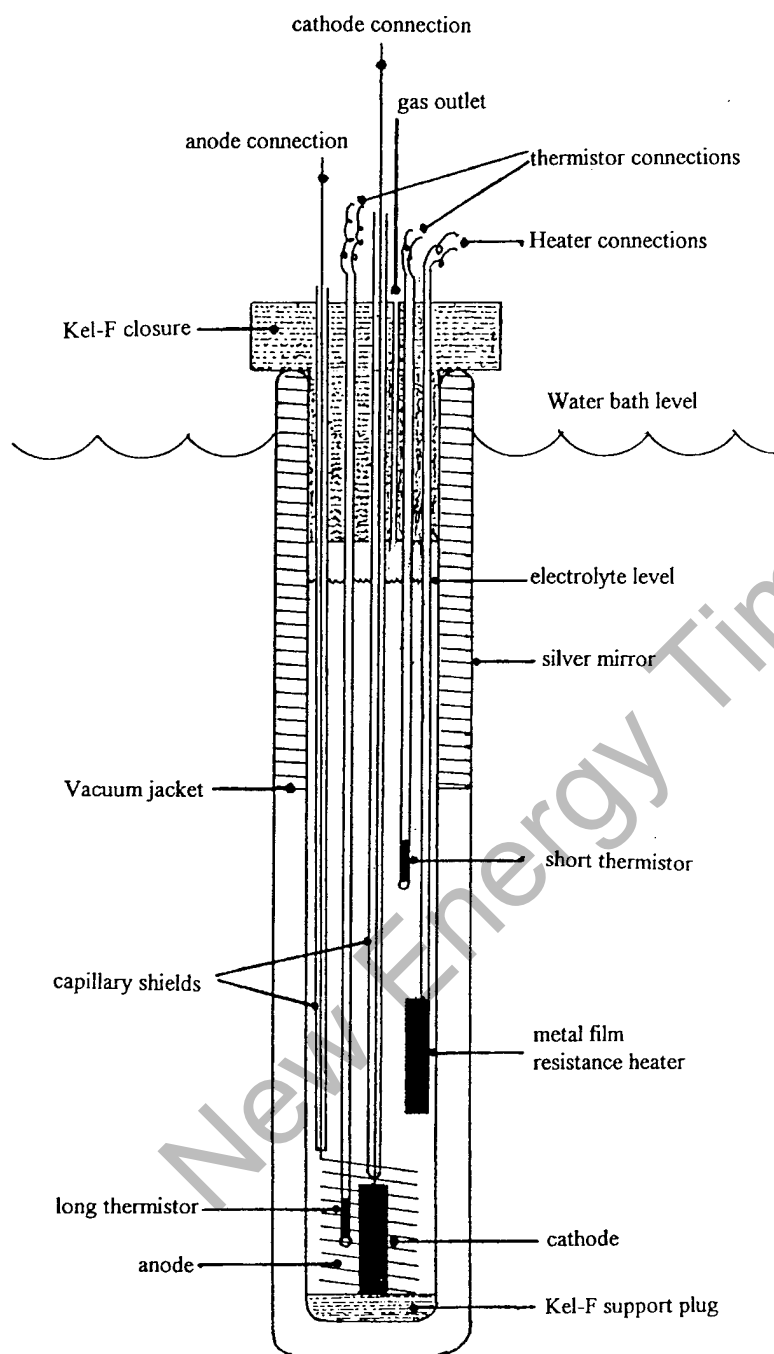


Fig. A.1 Schematic diagram of an ICARUS-1 Type Electrochemical Cell used in the Pd-B experiment. At NHE the long thermistor was positioned somewhat above the anode spiral. The NHE cell was 25 cm in length (with the top 8.0 cm silvered) and 2.5 cm in diameter.

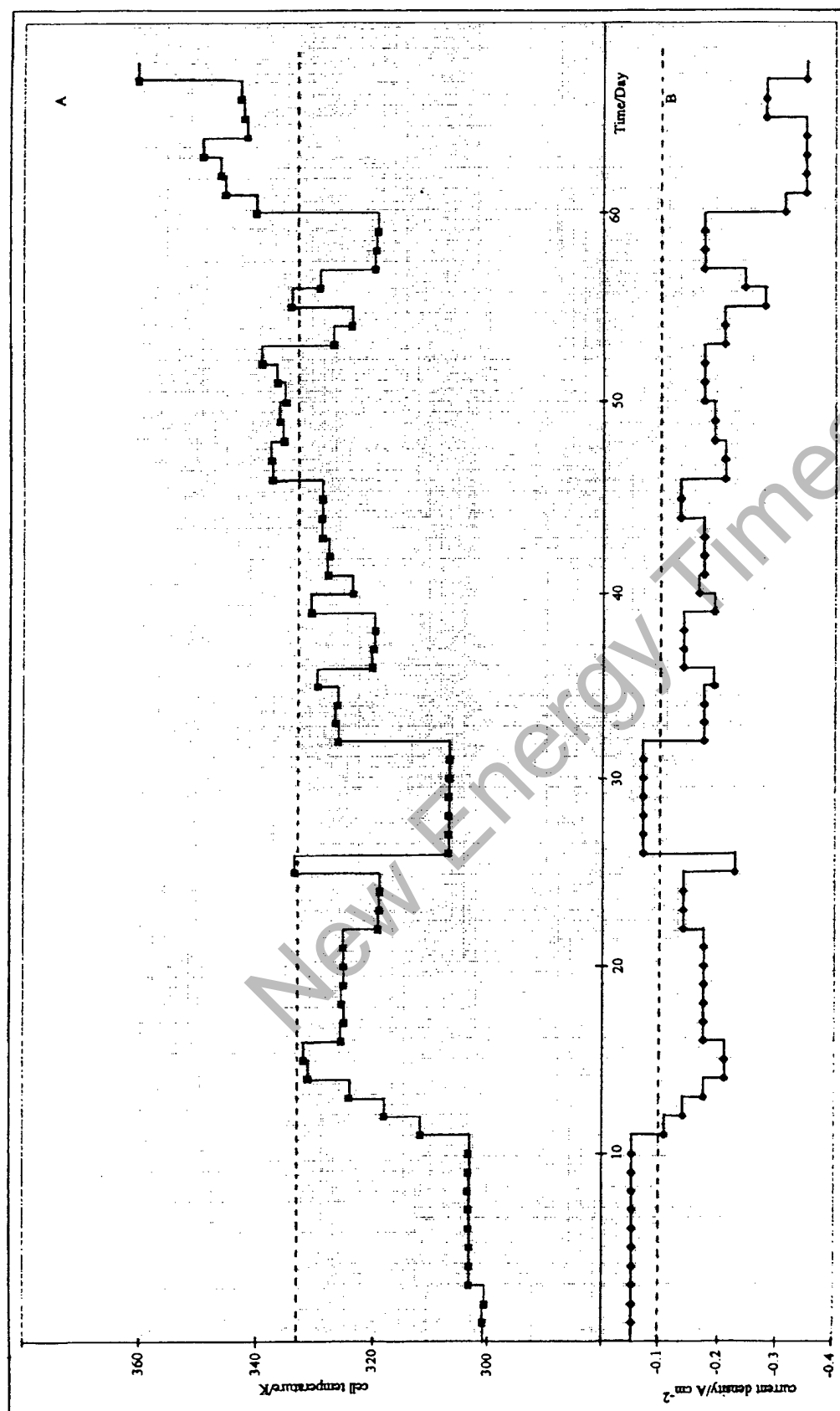


Fig. A.2 Variation of the cell temperature (A) and current density (B) for the Pd-B experiment. The dotted lines delineate the regions for the expected onset of significant levels of "positive feedback" (A) and excess enthalpy generation (B).

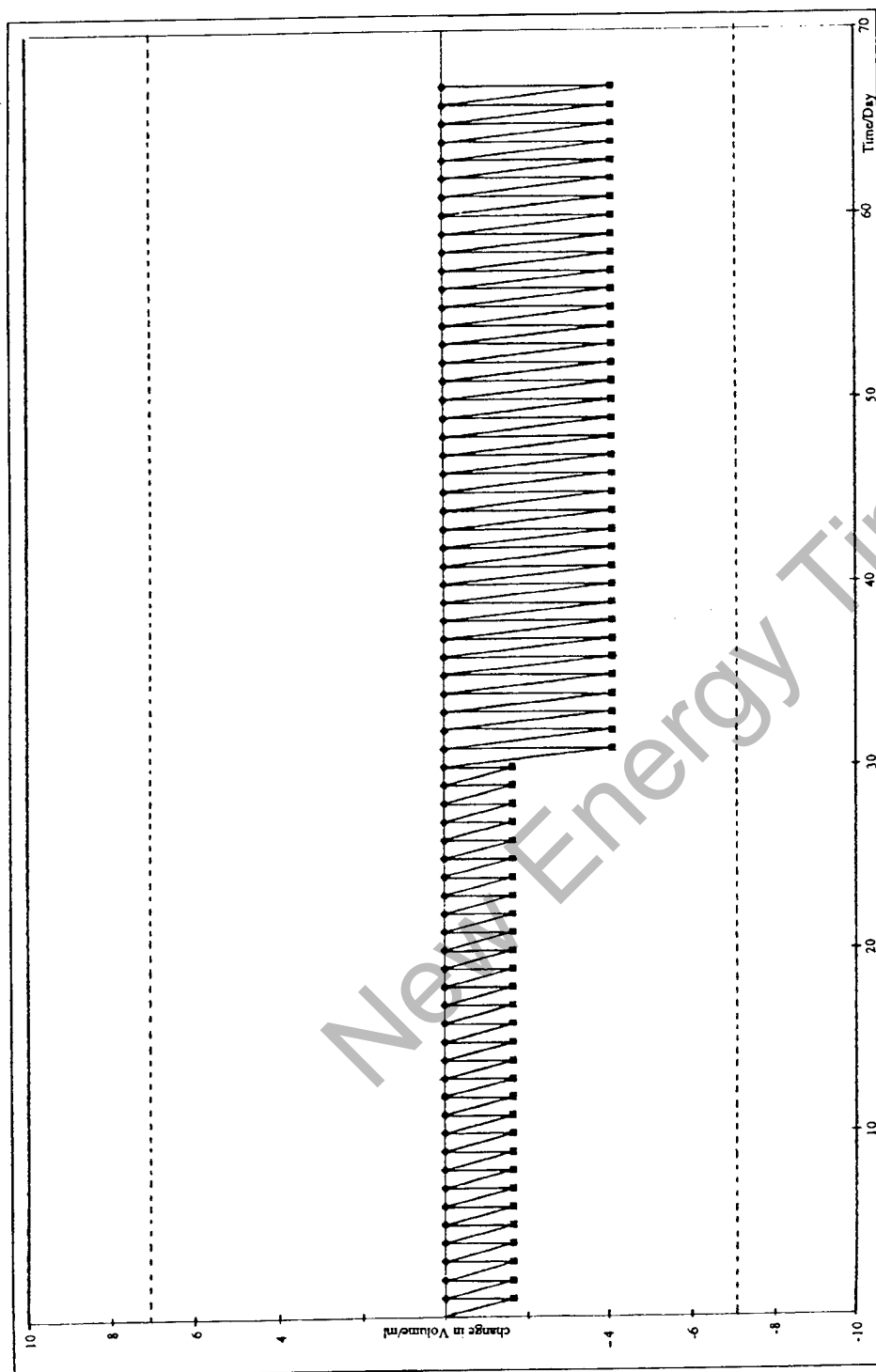


Fig. A.3 The changes in the volume of electrolyte in an ICARUS-1 Type Cell for an hypothetical experiment carried out according to the ICARUS-Systems Protocol but with measurement cycles lasting 1 day. The cell is assumed to have been replenished at the start of each measurement cycle. Cell current = 0.2 A for 29 days and 0.5 A for a further 40 days. The dotted lines delineate the changes in cell volume above which the level of electrolyte must be assumed to approach the base of the Kel-F plug sealing the top of the cell (Fig. A.1) and below which the level reaches the base of the top silvered portion of the Dewar-type cell.

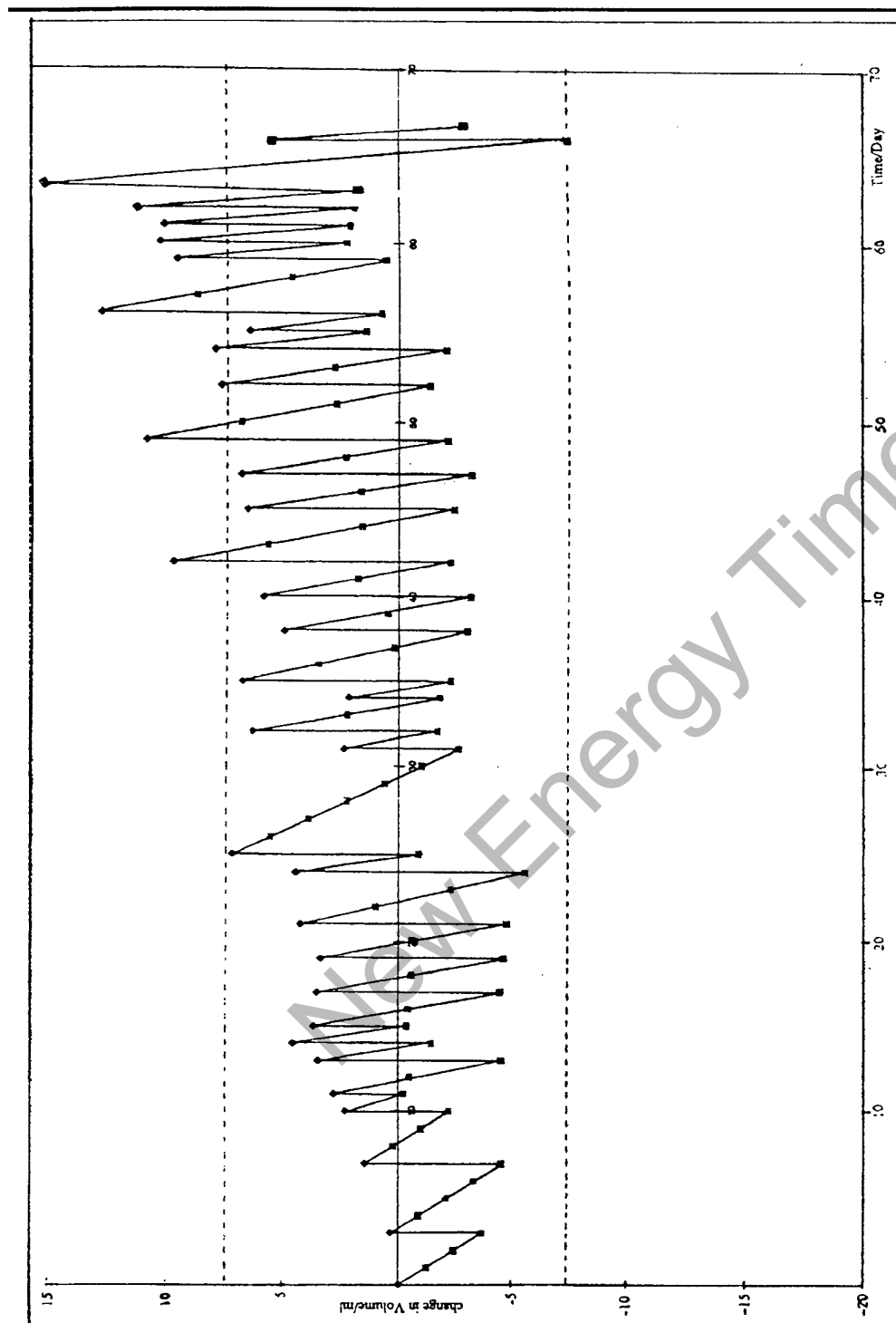


Fig A.4

The calculated changes in the volume of electrolyte in the ICARUS-1 Type Cell used in the Pd-B experiment assuming that the D_2O is removed from the cell only by electrolysis (no evaporative loss). The polarizations were carried out at the current densities shown in Fig. A.2B. The dotted lines delineate the changes in cell volume above which the level of electrolyte must be assumed to approach the base of the Kel-F plug sealing the top of the cell (Fig. A.1) and below which the level reaches the base of the silvered portion of the Dewar-type cell. Experimental measurements of the electrolyte volume show that the volume of D_2O lost by electrolysis should be multiplied by 1.04 to correct for the evaporative loss of D_2O .

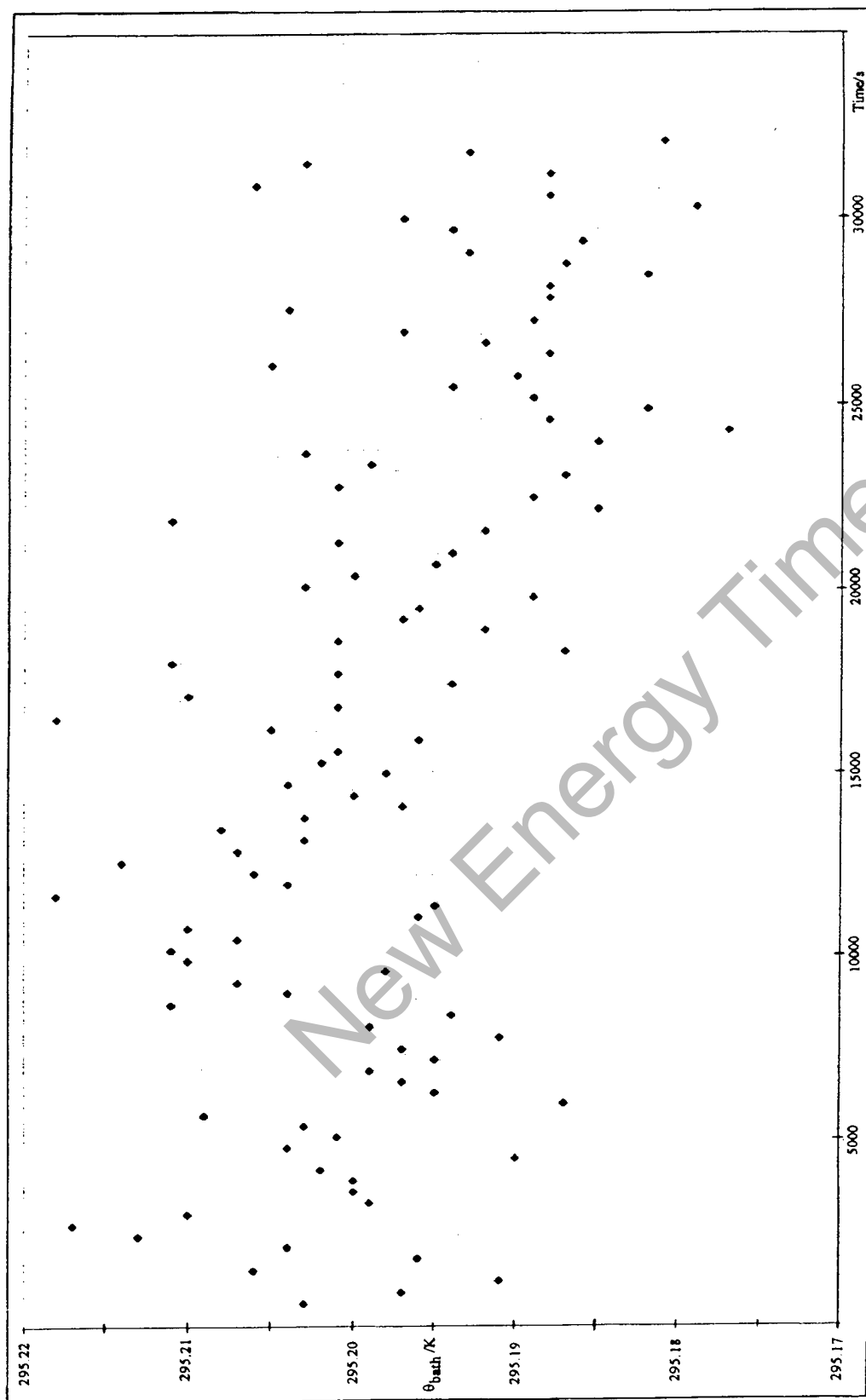


Fig. A.5 The temperature of the water bath for the first 32,400 s of the first measurement cycle (i.e. the period $0 < t < t_1$).

Mean temperature = 295.198 K

σ (temperature difference) = 0.0088 K

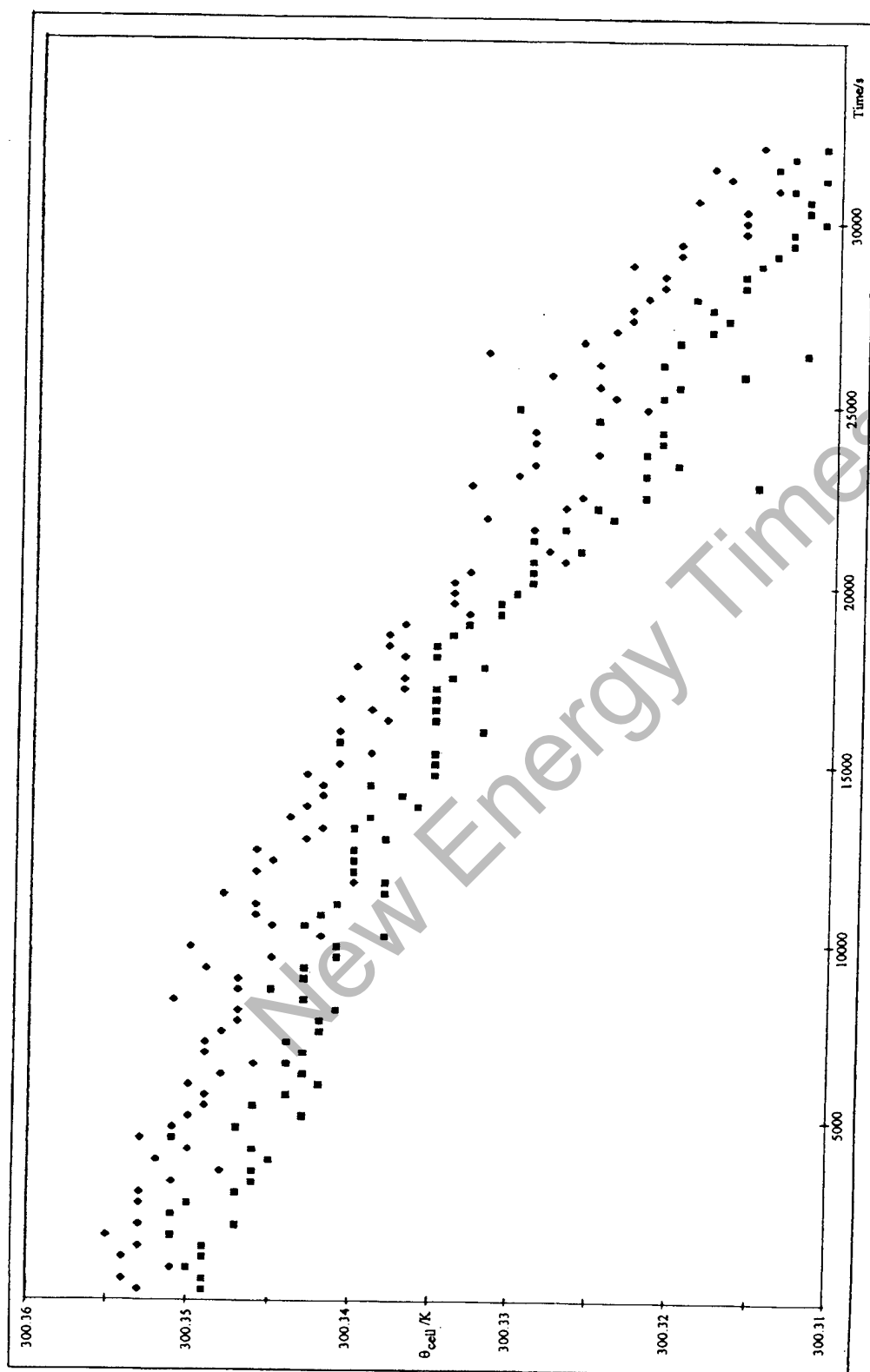


Fig. A.6 The cell temperature at the two measurement locations for the first 32,400 s of the first measurement cycle (i.e. the period $0 < t < t_1$). Mean temperature difference = 0.0045 K; σ (temperature difference) = 0.0027 K

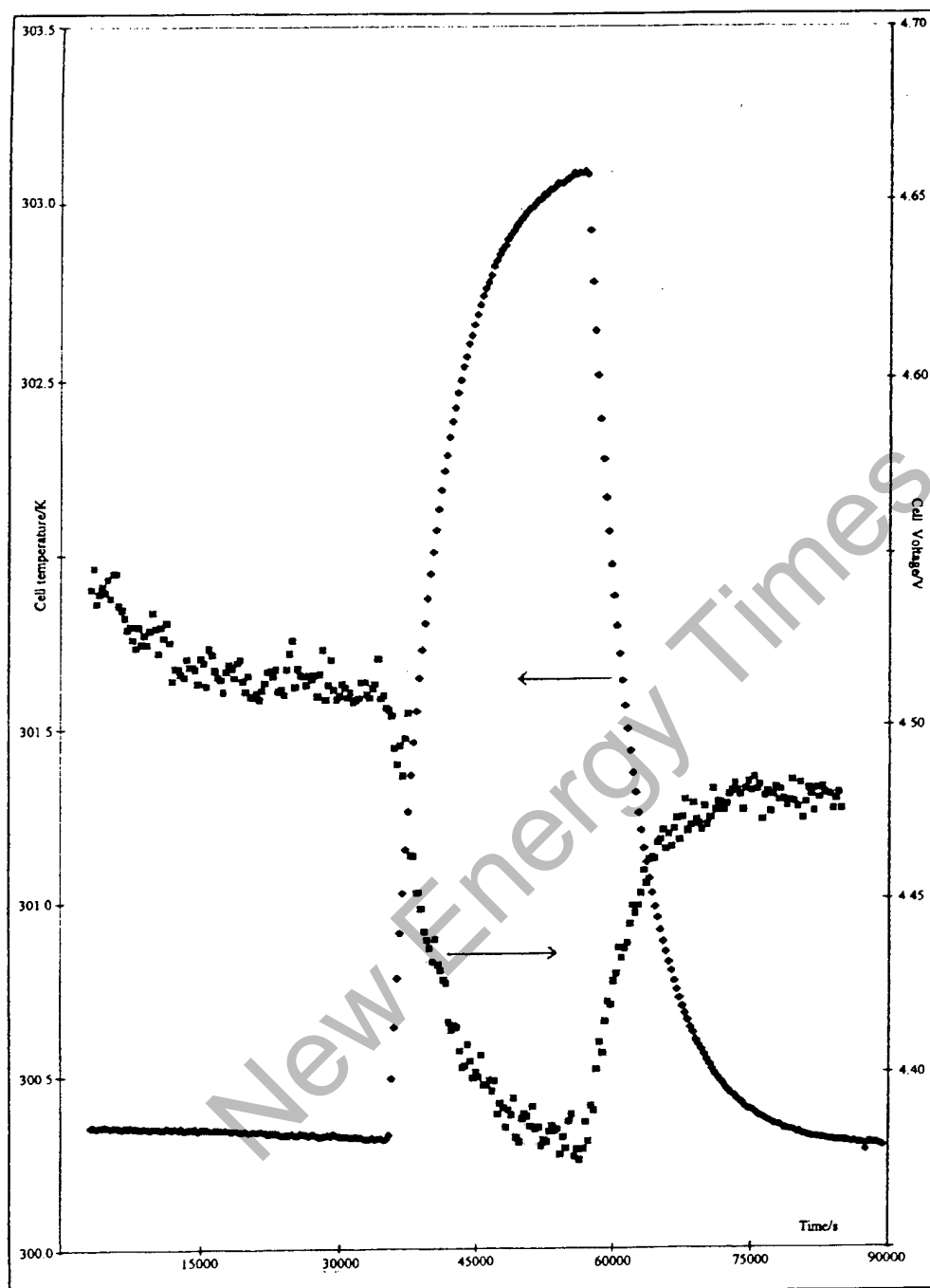


Fig. A.7 The "raw data" (the cell temperature and cell potential as a function of time) for the third measurement cycle of the Pd-B experiment.

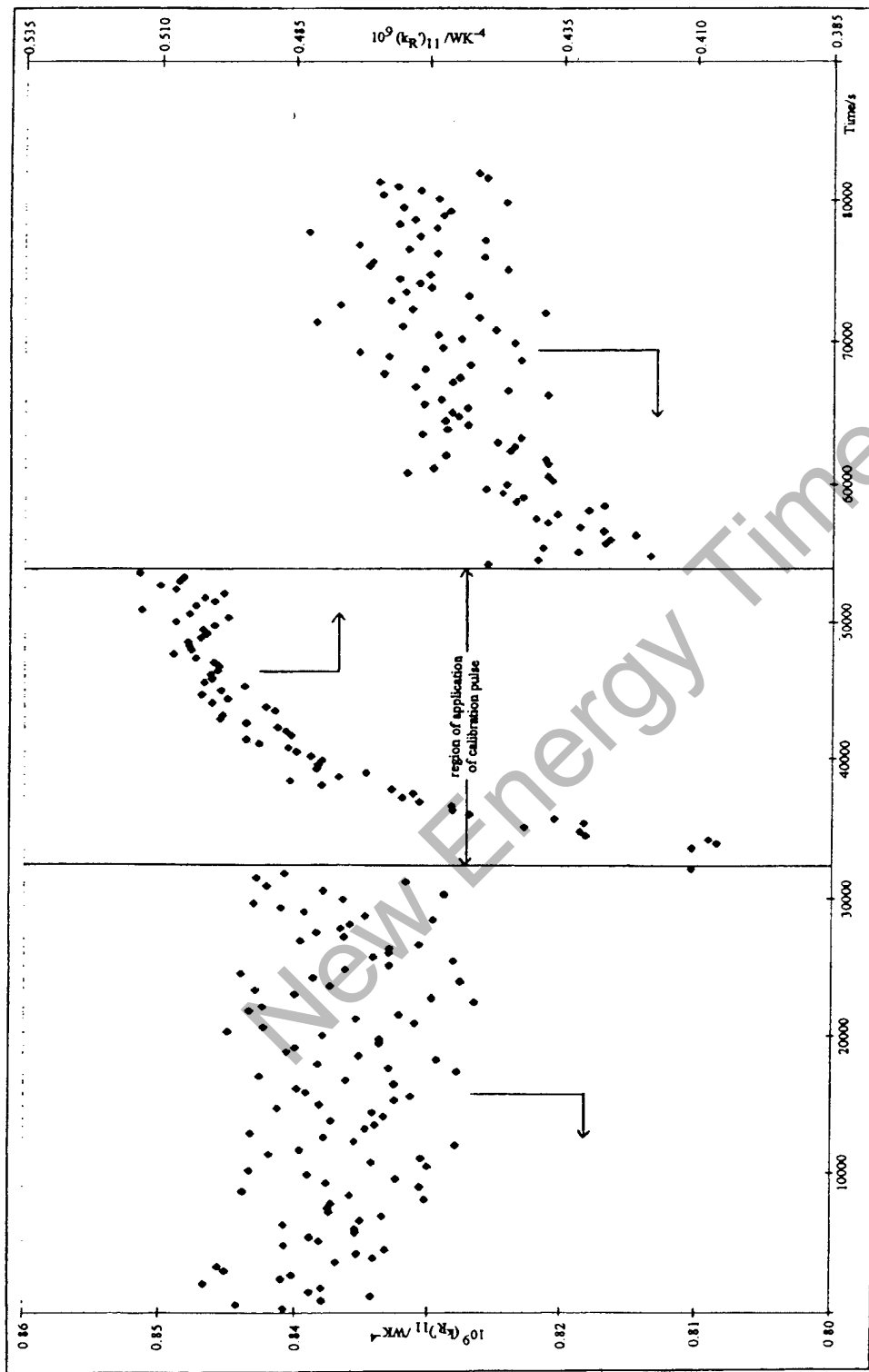


Fig. A.8 The "lower bound heat transfer coefficient, $(k'_R)_{11}$ ", as a function of time for the third measurement cycle as determined by the analysis provided by the group at the NHE Laboratories. The vertical lines delineate the period of application of the calibration pulse, $(t_1 < t < t_2)$. The amplitude of the calibration pulse, $\Delta Q \approx 0.2500 \text{ W}$, has been excluded in the calculation of $(k'_R)_{11}$ during the period $t_1 < t < t_2$ and it has been assumed that $C_p M = 490 \text{ WK}^{-1}$

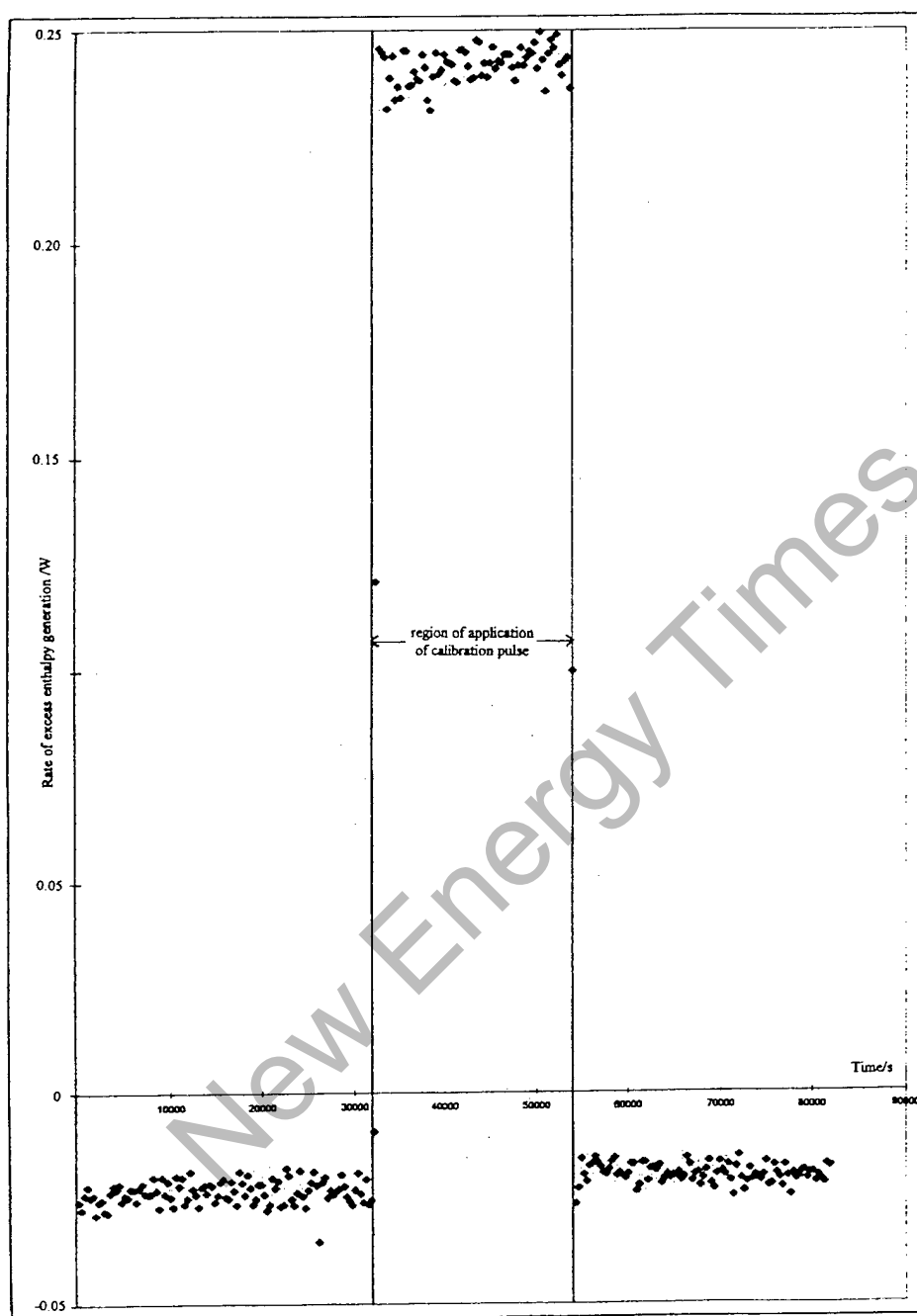


Fig. A.9 The rate of excess enthalpy generation Q/W , as a function of time for the third measurement cycle as determined in the analysis provided by the group at the NHE Laboratory.

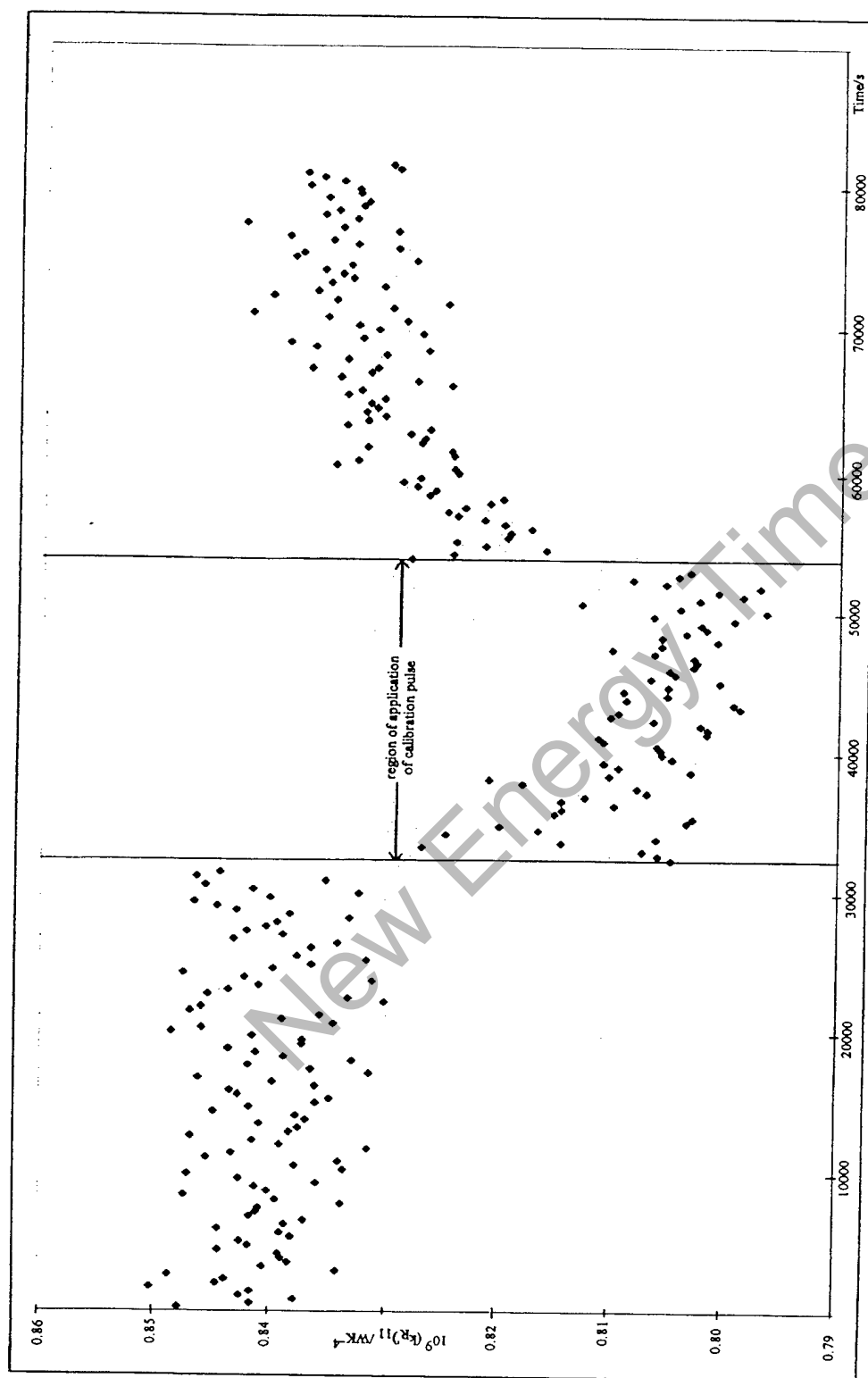


Fig. A.10 The "lower bound heat transfer coefficient, $(k'_R)_{11}$ " as a function of time for the third cycle as determined by the ICARUS-Systems procedure with the inclusion of the calibration pulse, $\Delta Q = 0.2500$ W, in the calculation of $(k'_R)_{11}$ during the period $t_1 < t < t_2$. It was assumed that $C_p M = 490$ JK⁻¹.

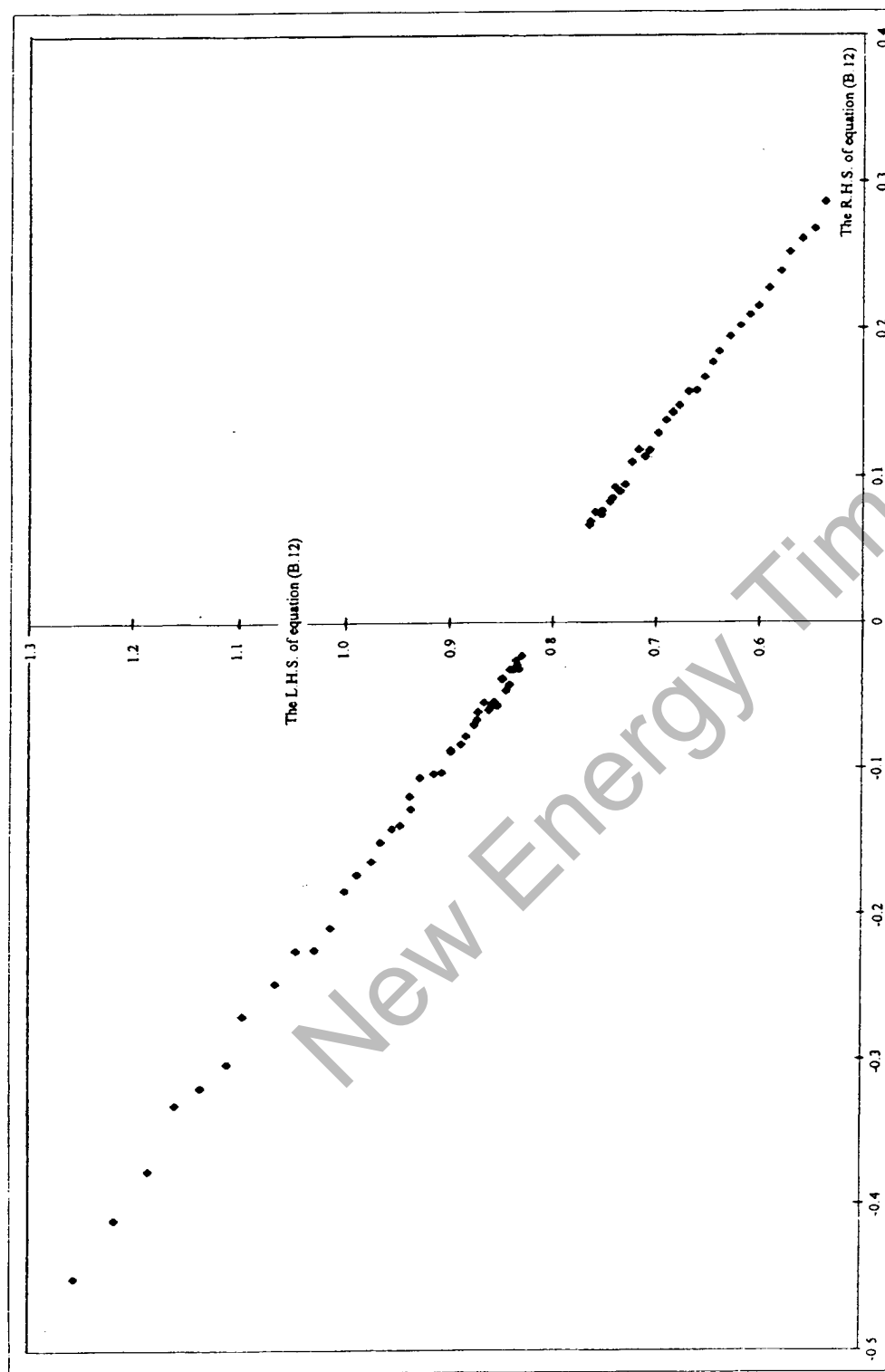


Fig. A.11 Evaluation of $(k'_R)_{181}$ and C_{pM} according to equation (B.12). The values derived and the relevant regression coefficients are shown in Column 6 of Table A.3 according to the range of the ordinates and abscissae used to derive the straight line fits.

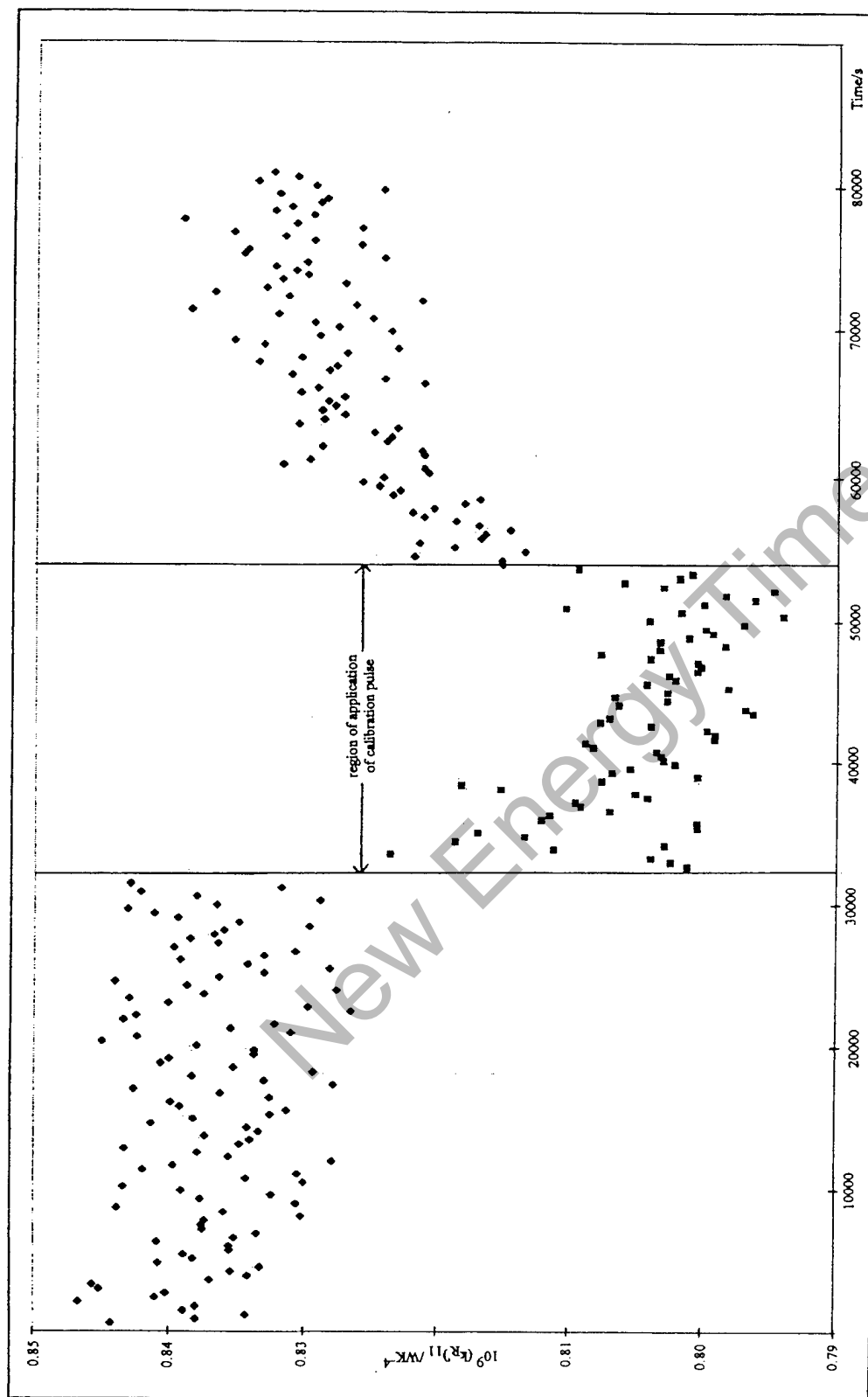


Fig. A.12 The "lower bound heat transfer coefficient, $(k_R')_{II}$ " as a function of time for the third measurement cycle as determined by the ICARUS-Systems procedure with the inclusion of the calibration pulse, $\Delta Q = 0.2500$ W, in the calculation of $(k_R')_{II}$ during the period $t_1 < t < t_2$ and with all remaining corrections to the calculation (see the main text). It has been assumed that $C_p M = 490 \text{ JK}^{-1}$.

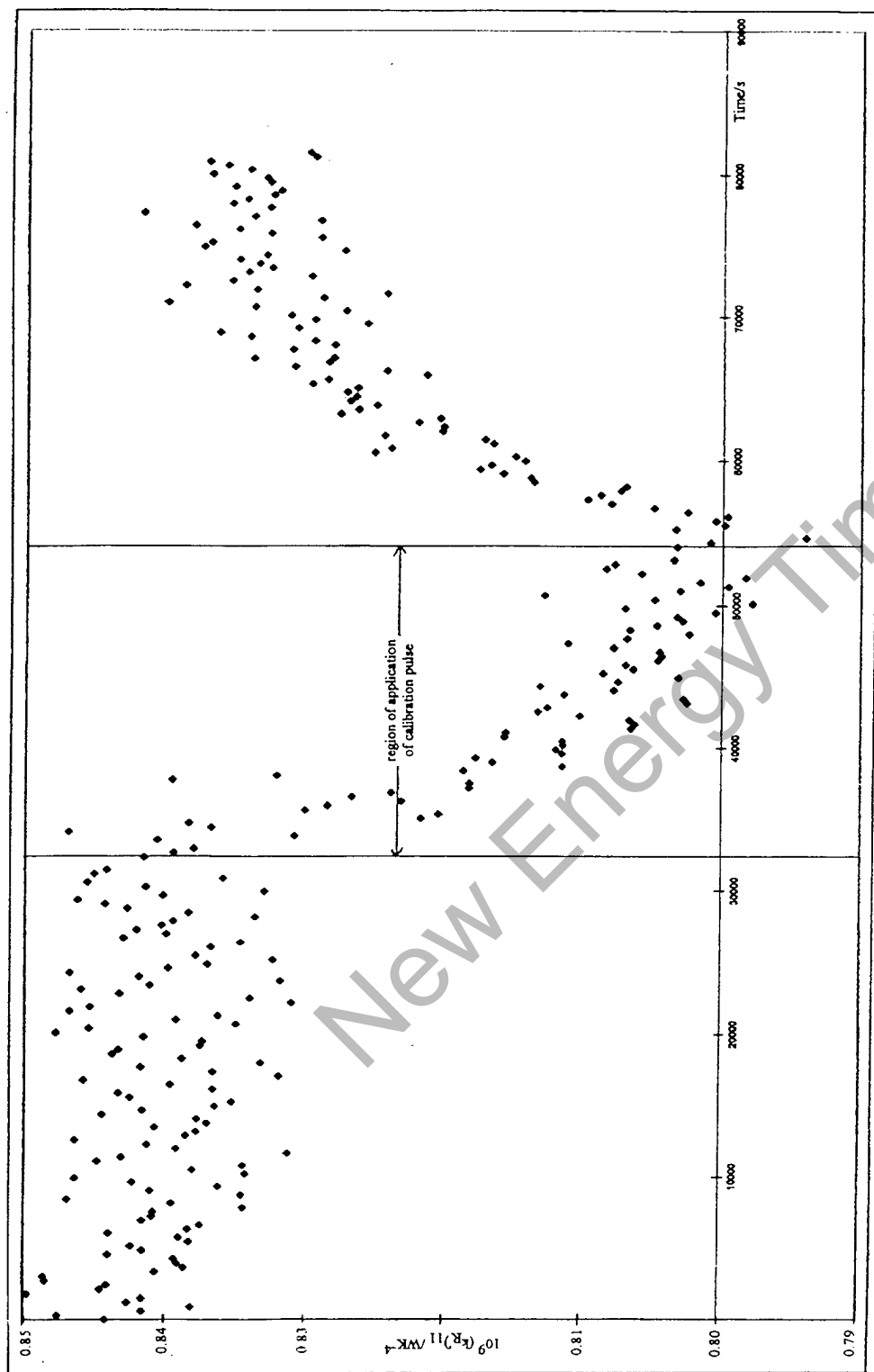


Fig. A.13 The "lower bound heat transfer coefficient, $(k'_R)_{11}$ " as a function of time for the third cycle determined by the ICARUS-Systems procedure with the inclusion of the calibration pulse, $\Delta Q = 0.2500$ W, in the calculation of $(k'_R)_{11}$ during the period $t_1 < t < t_2$ and with all remaining corrections to the calculation (see the main text). It has been assumed that $C_p M = 450$ JK⁻¹.

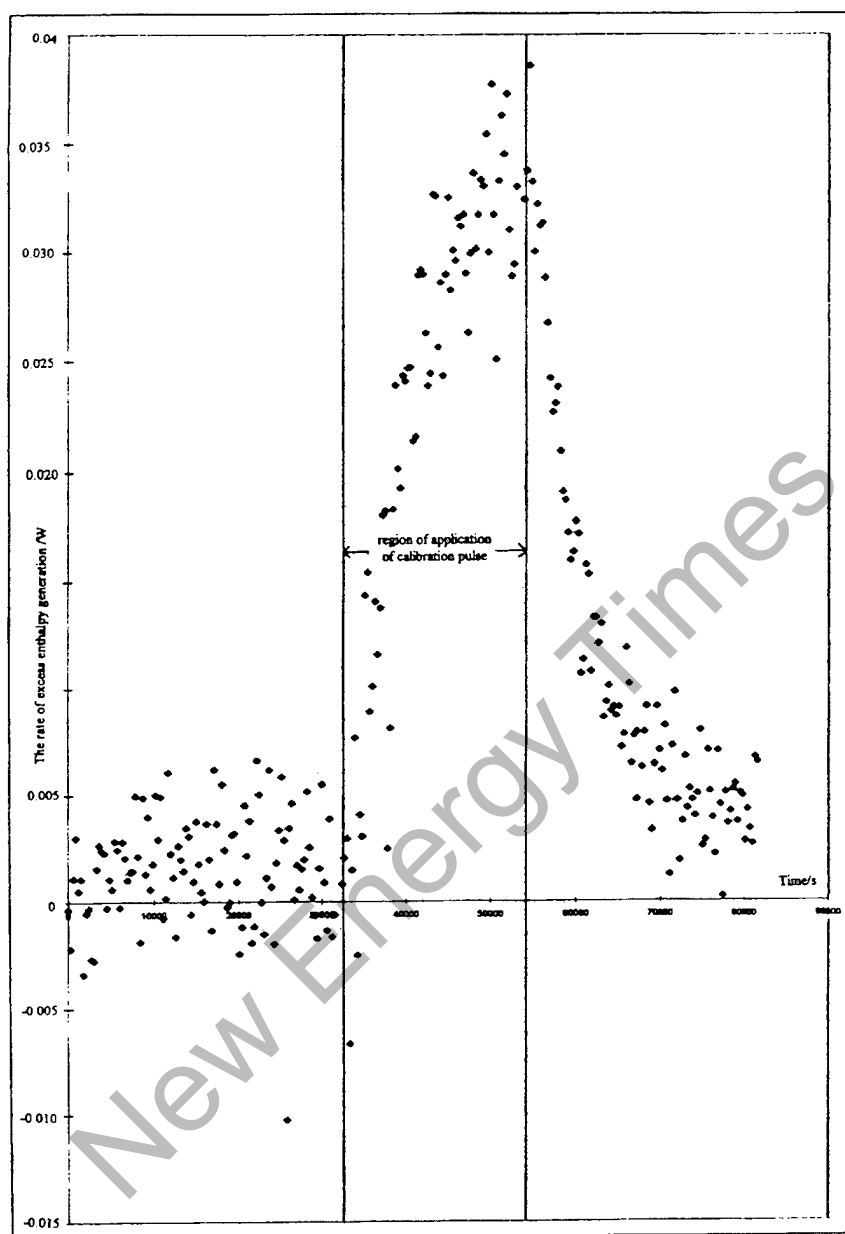


Fig. A.14 The rate of excess enthalpy generation, Q , as a function of time for the third measurement cycle as determined by the ICARUS-Systems procedure using the "true heat transfer coefficient, $(k'_R)_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ " and the values for the "lower bound heat transfer coefficient, $(k'_R)_{11}$ " shown in Fig. A.12.

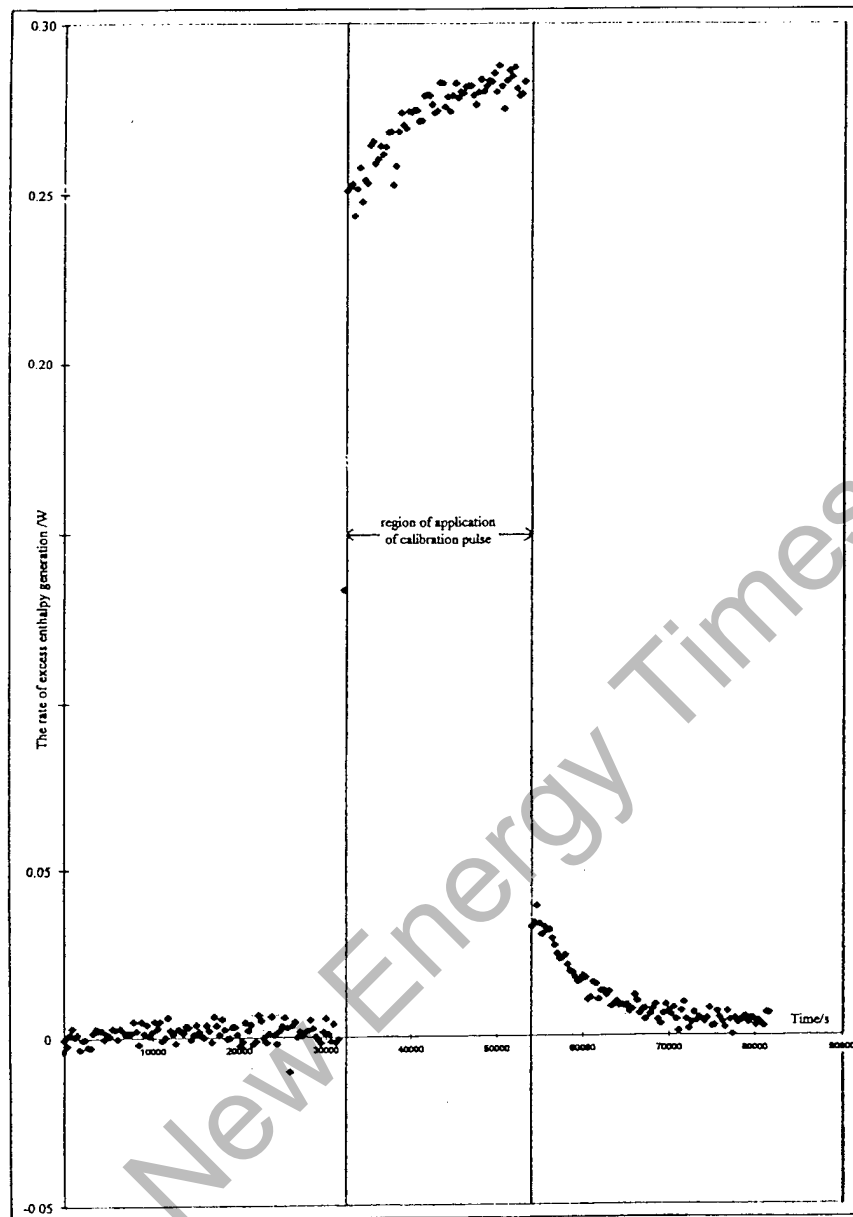


Fig. A.15 The rate of excess enthalpy generation, Q , as a function of time for the third measurement cycle determined using the "true heat transfer coefficient, $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$. The values of the "lower bound heat transfer coefficient, $(k_R')_{11}$," have been calculated as in the ICARUS-Systems procedure (see Fig. A.12) but with the exclusion of the calibration pulse, $\Delta Q = 0.2500 \text{ W}$, during the period $t_1 < t < t_2$ (c.f. the NHE methodology).

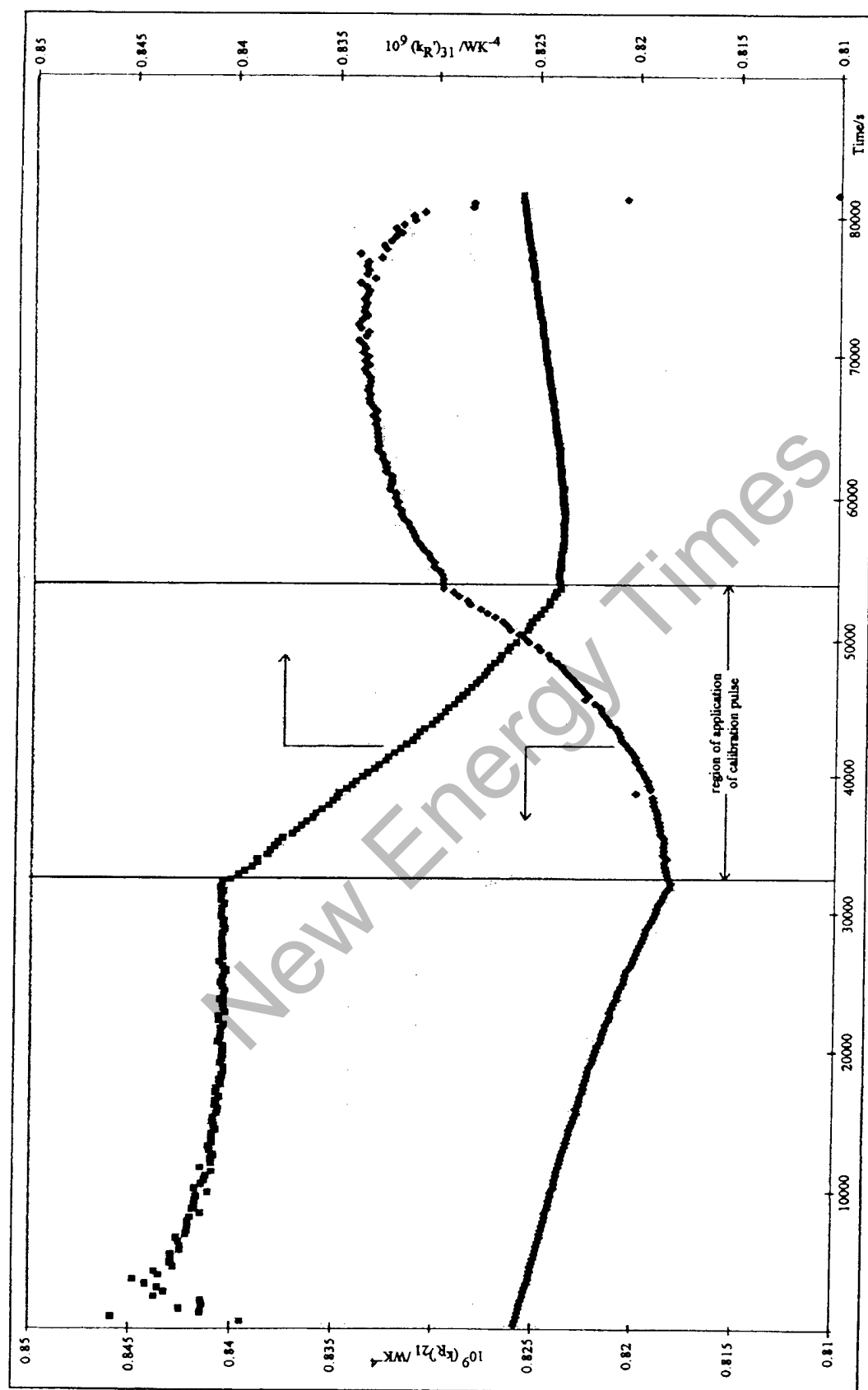


Fig. A.16 The variation of $(k_R')_{21}$ and $(k_R')_{31}$ with time for the whole of the third measurement cycle of this Pd-B experiment.

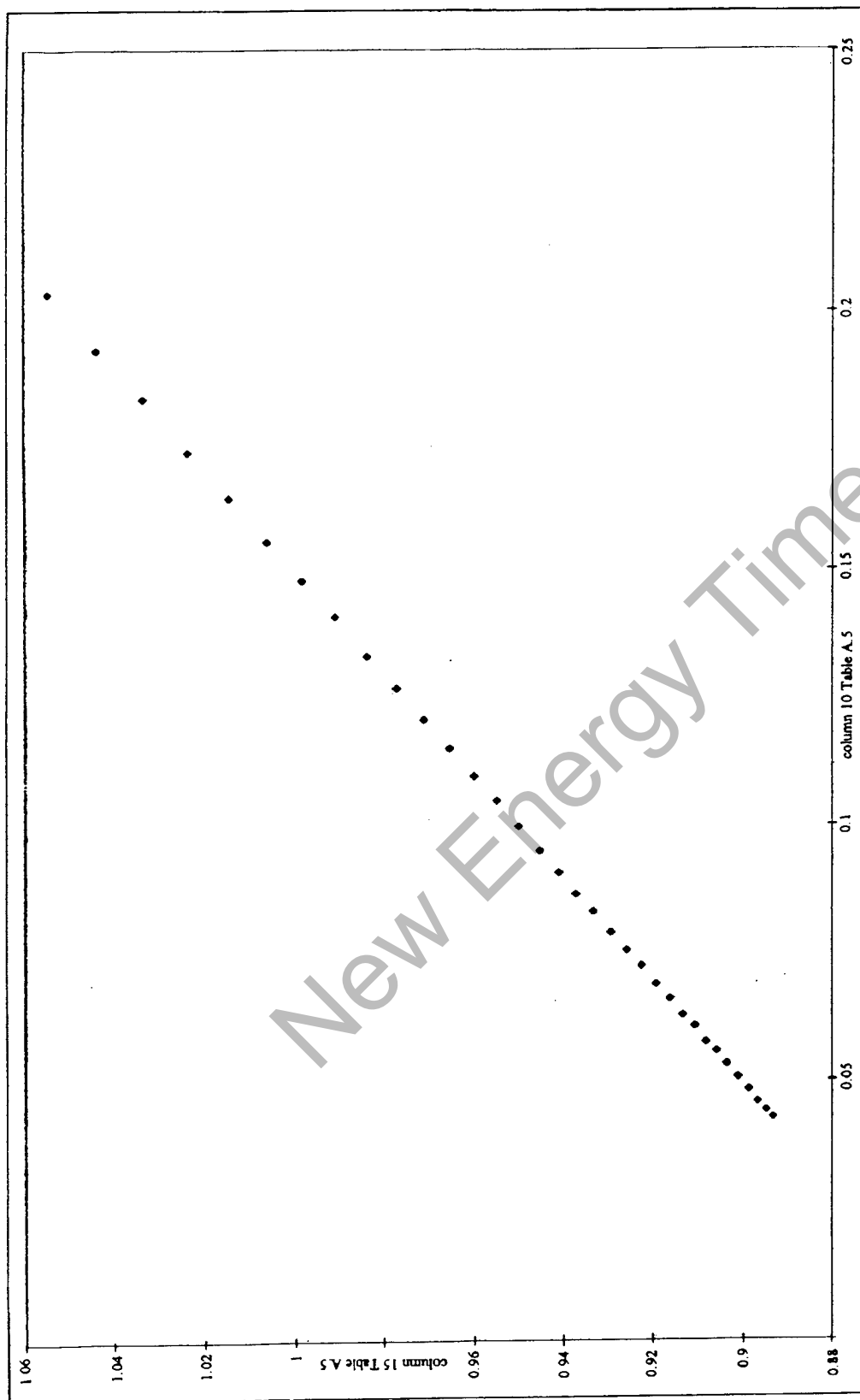


Fig. A.17 Evaluation of the integral heat transfer coefficient $10^9 (k'_R)_{262}^{-1} / \text{WK}$ and water equivalent C_{pM} / JK^{-1} for the third measurement cycle of this experiment with correction for the effects of "positive feedback" (see Columns 10, 14 and 16 of Table A.5).

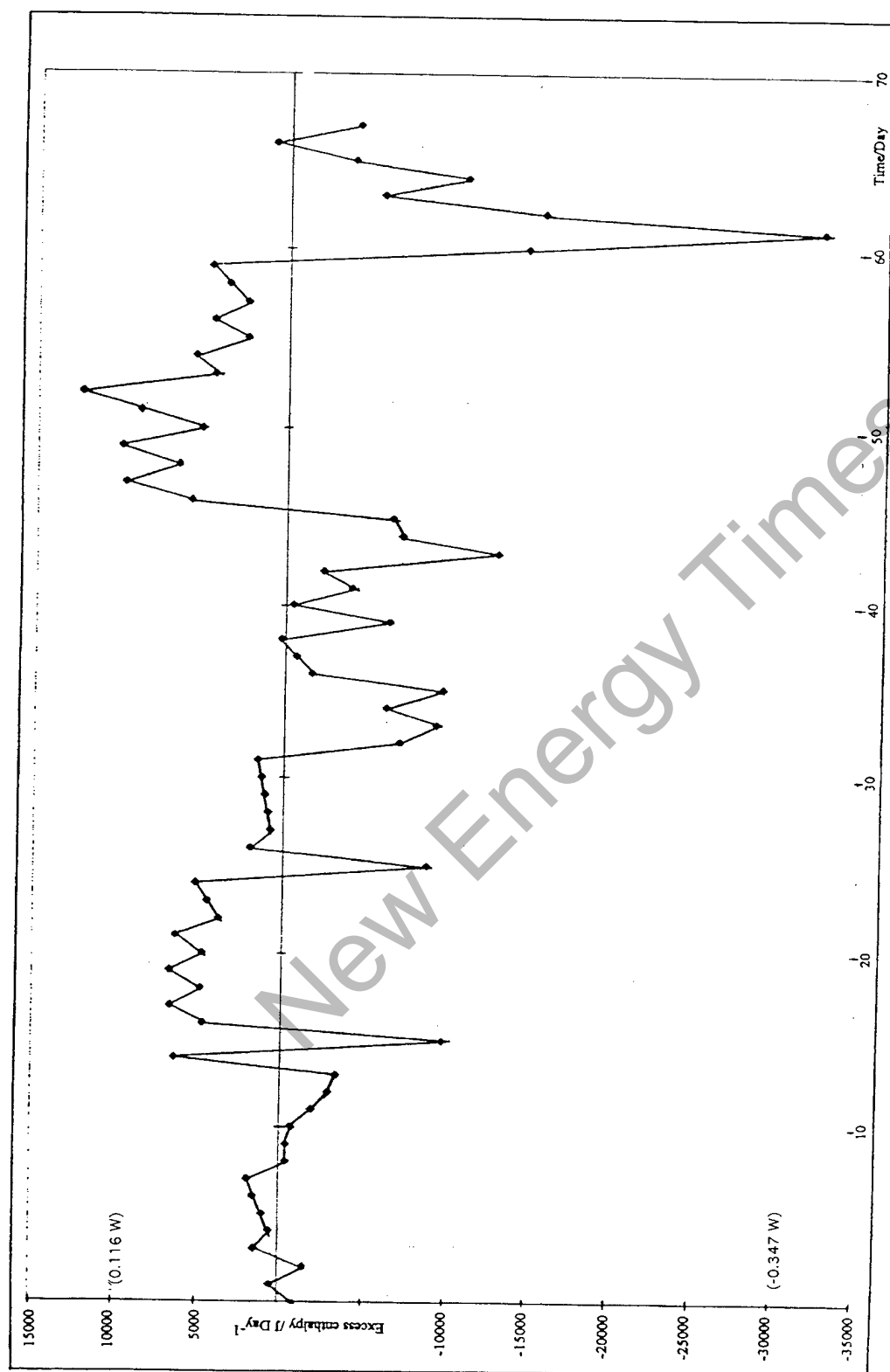


Fig. A.18 The excess enthalpy as a function of time for this Pd-B experiment evaluated using the "true heat transfer coefficient, $(k_R)'_{12} = 0.79350 \times 10^{-9} \text{ WK}^{-4}$ " as given by the analysis of the group at the NHE Laboratory.

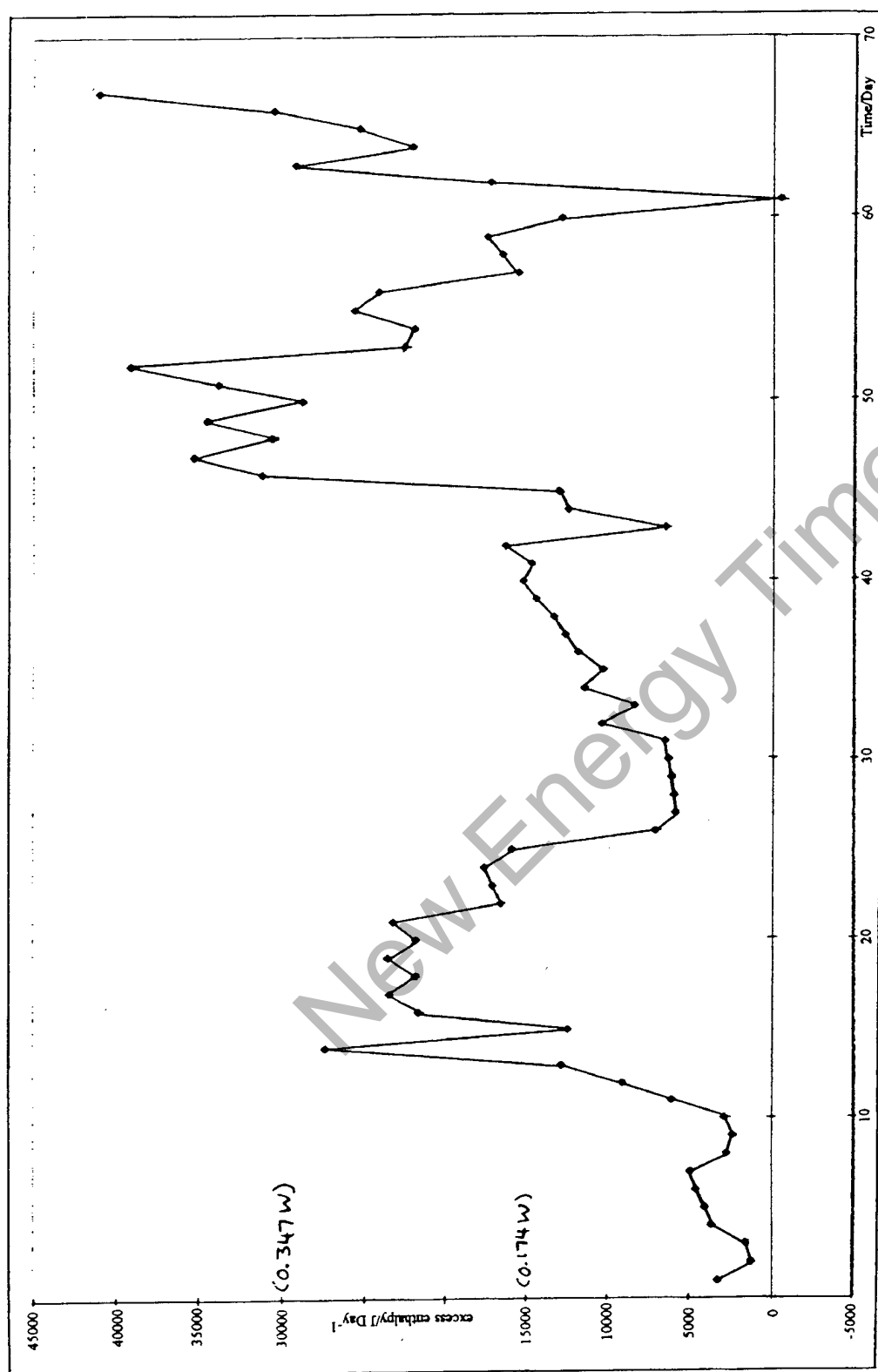


Fig. A.19 The excess enthalpy as a function of time for this experiment evaluated using the "true heat transfer coefficient,

$(k_r')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ as given by the ICARUS-Systems analysis modified to take account of "positive feedback".

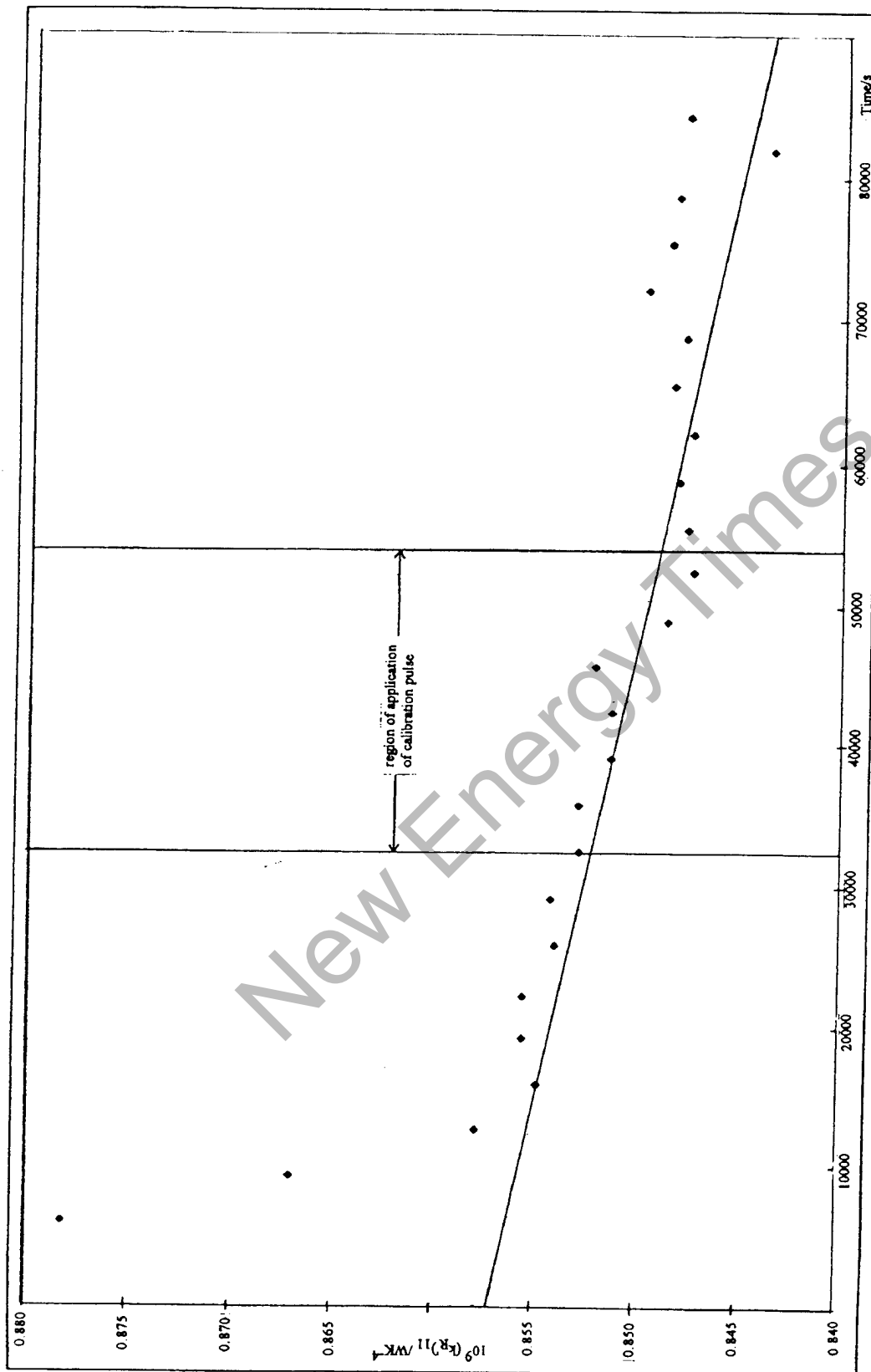


Fig. A.20 The variation with time of the 11-point average of the "lower bound heat transfer coefficient, $10^9 (k_R)_{11}$ " for Day 61 of this experiment. The full line gives the variation with time of $10^9 (k_R)_{11}$ determined for relevant "blank" experiments ^{(4),(13),(14)}

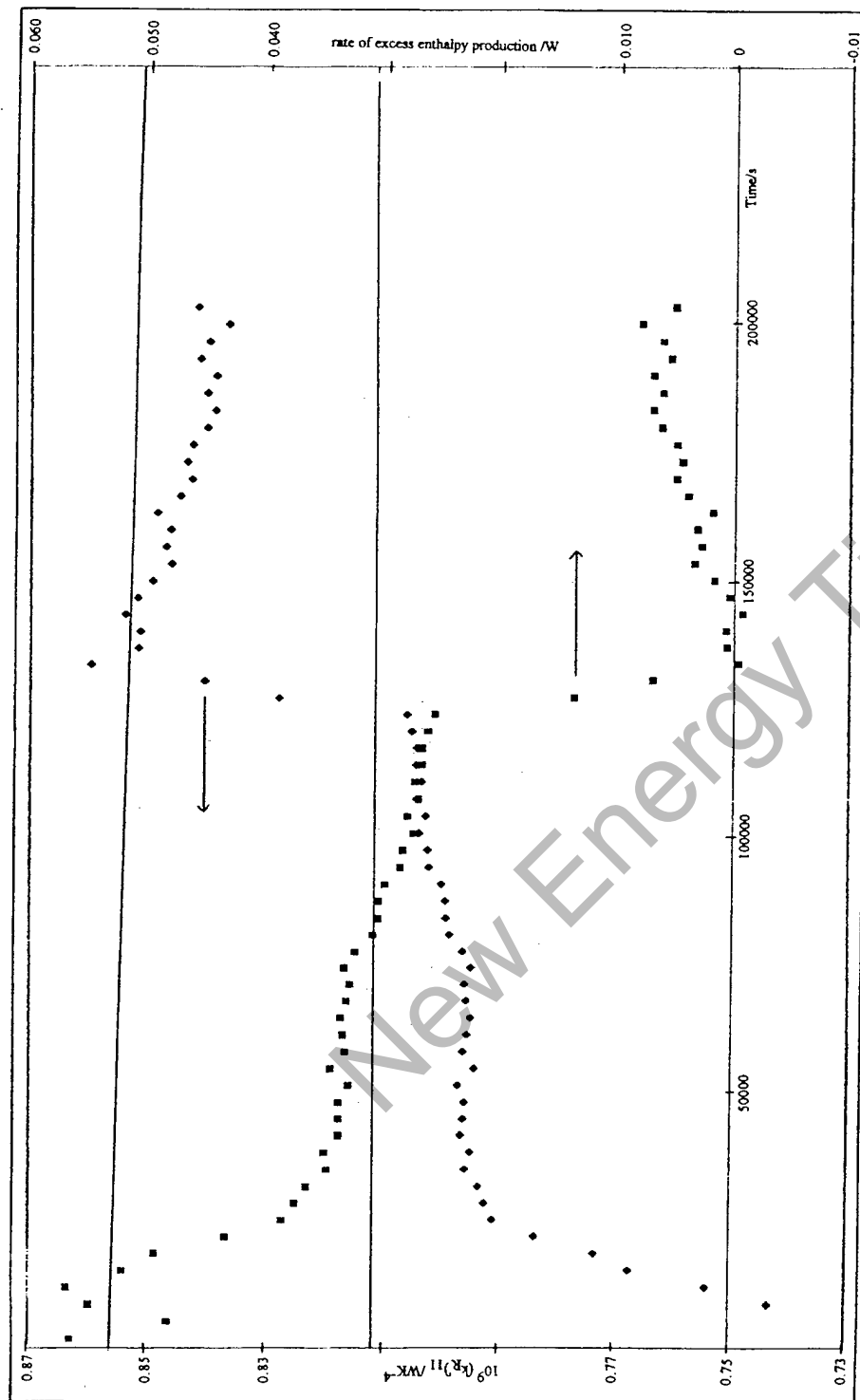


Fig. A.21 The "lower bound heat transfer coefficient, $(k_R)_{11}$ " and the rate of excess enthalpy generation, Q , for the first 57 hours of operation of this experiment. The full line at the top shows the expected variation of $(k_R)_{11}$ with time. Values of Q are based on the assumption that the "true heat transfer coefficient, $(k_R)_{12}$ " is given by the regression line. The horizontal line shows the value of Q based on the assumptions that the current efficiency for the charging of the electrode is 100% and that the heat of absorption of deuterium in the lattice is 40 kJ Mole^{-1} .

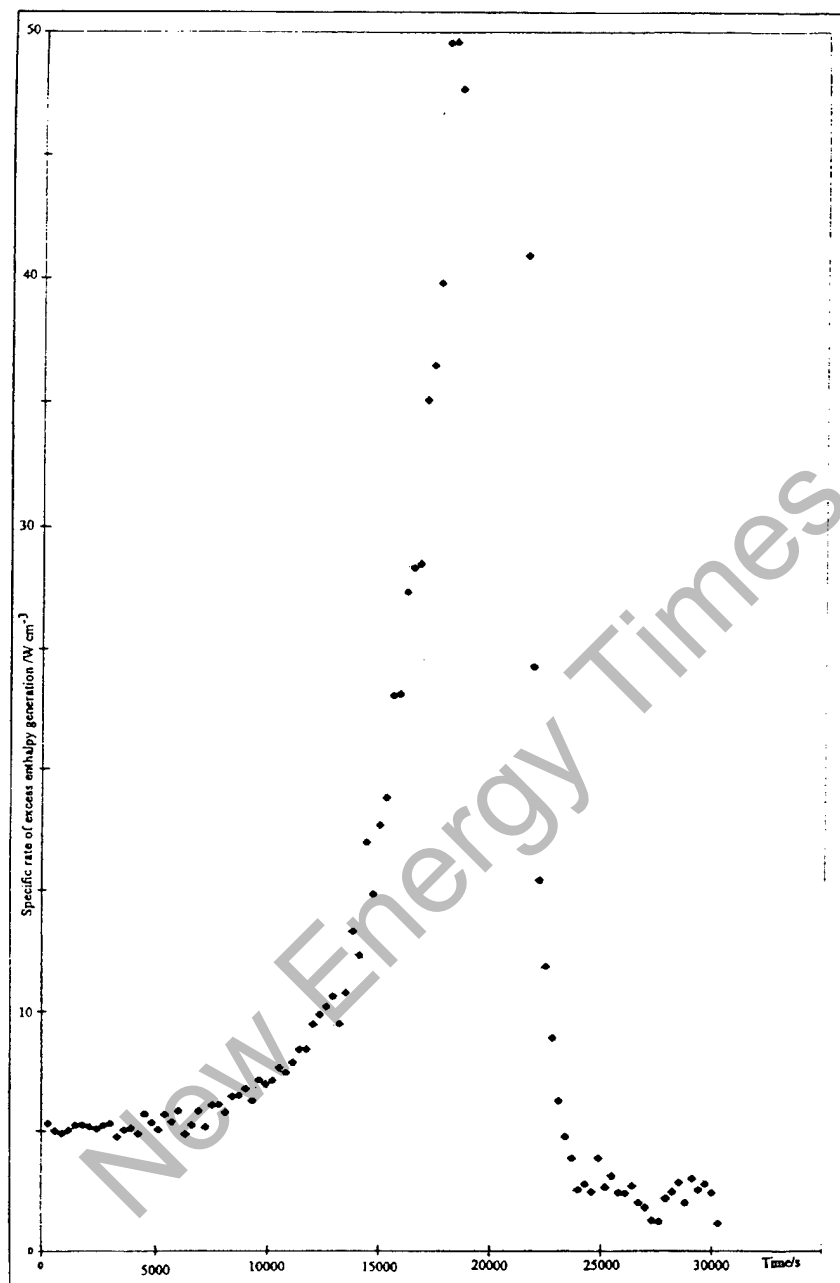


Fig. A.22 Comparison of the specific rates of excess enthalpy generation/ W cm^{-3} on day 68 of this experiment during the period $0 < t < 21,300 \text{ s}$ (where the cell is driven to dryness) and the initial period $21,300 \text{ s} < t < 30,300 \text{ s}$ of observation of "Heat after Death". [Due to correction for electrode dimensions, the values for W/cm^3 should be multiplied by a factor 0.54].

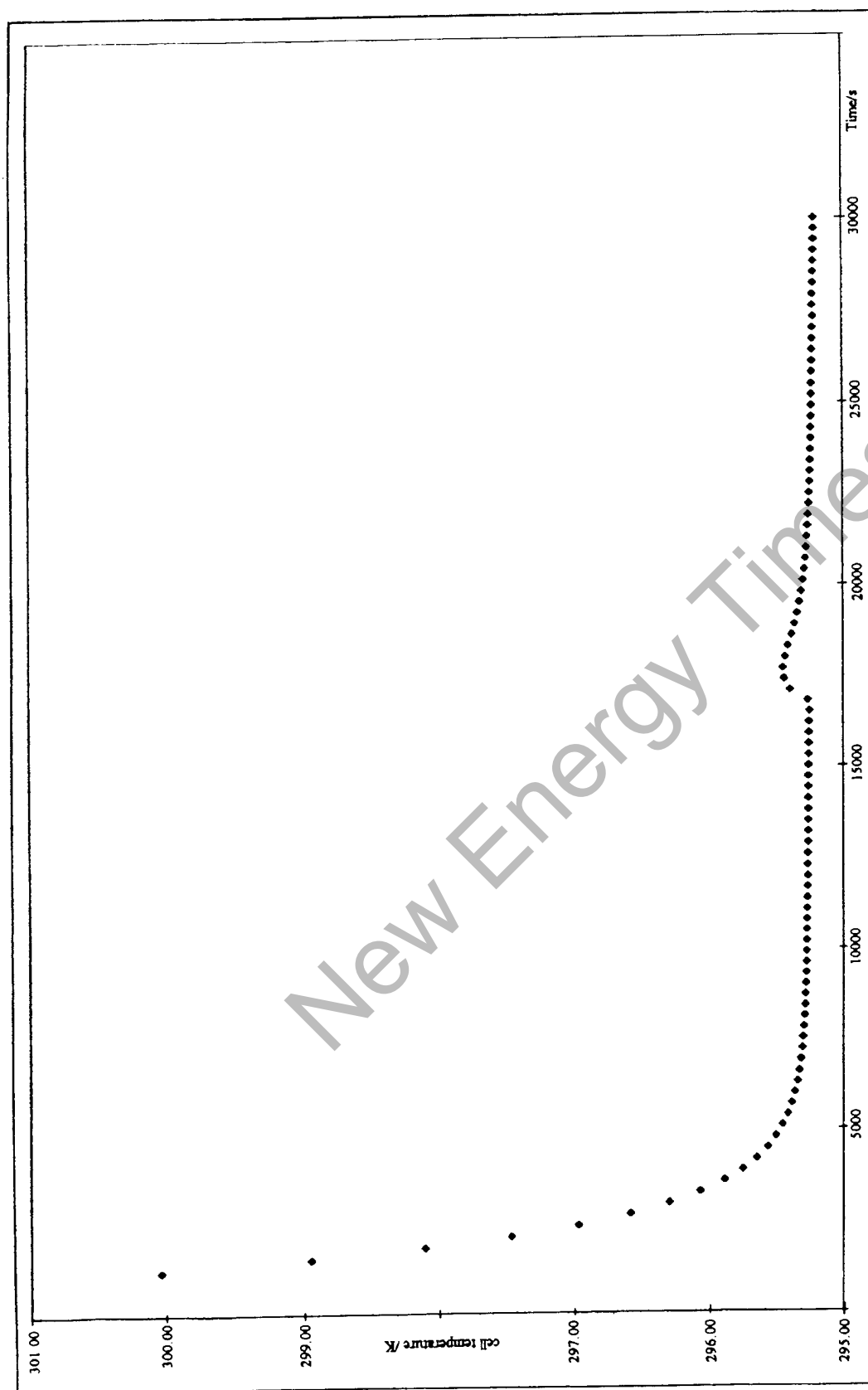


Fig. A.23 The "cooling curve" observed on Day 69 of the operation of this experiment following the disconnection of the cell from the galvanostat.

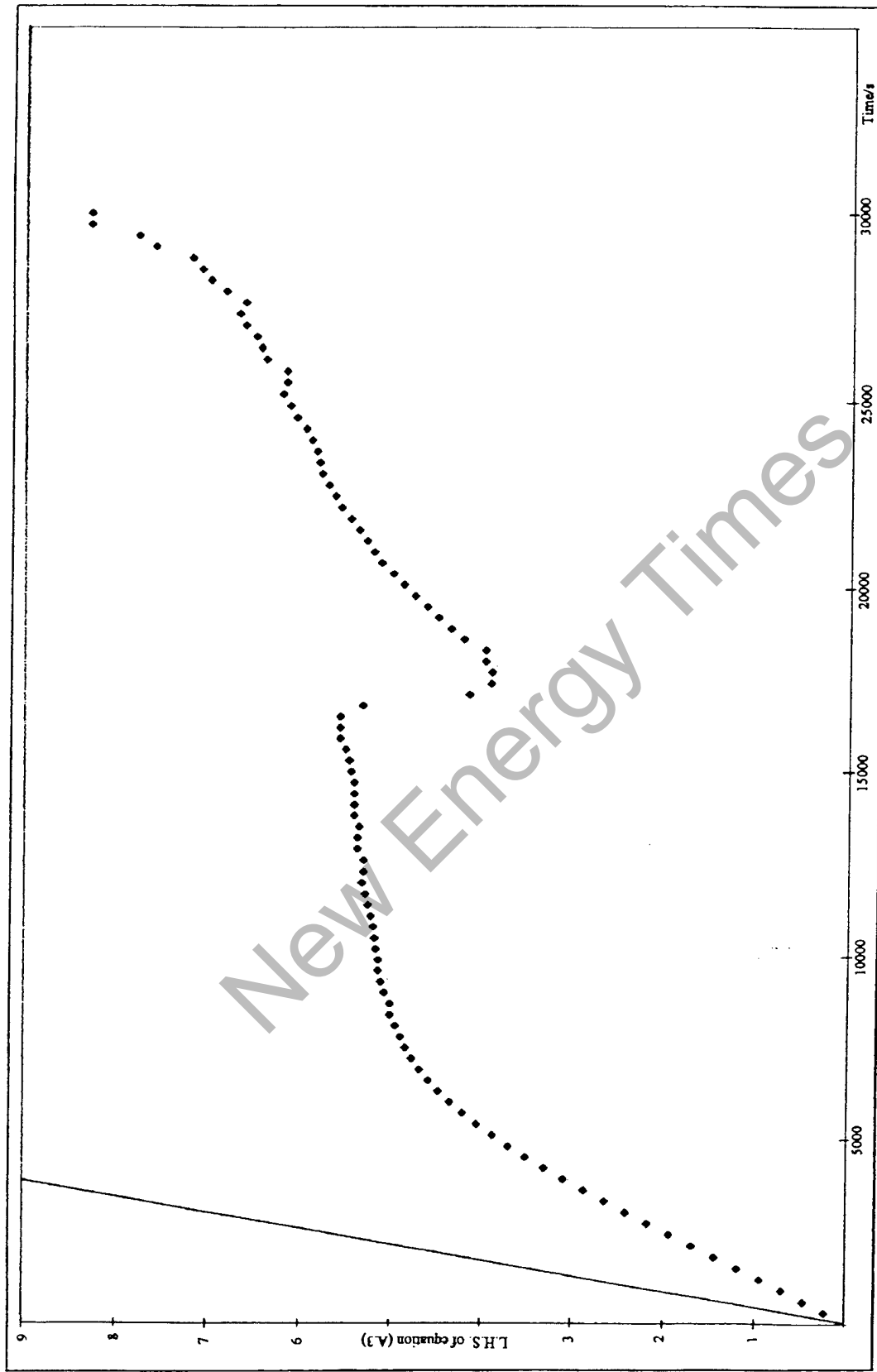


Fig. A.24 The analysis of the initial portion of the "cooling curve" shown in Fig. A.23 according to equation (A.3). The full line shows the R.H.S. of this equation plotted with $C_p M = 28.3 \text{ JK}^{-1}$, $(k_R')_{12} = 0.65 \times 10^{-9} \text{ WK}^{-4}$ and $\theta_{\text{bath}} = 295.204 \text{ K}$

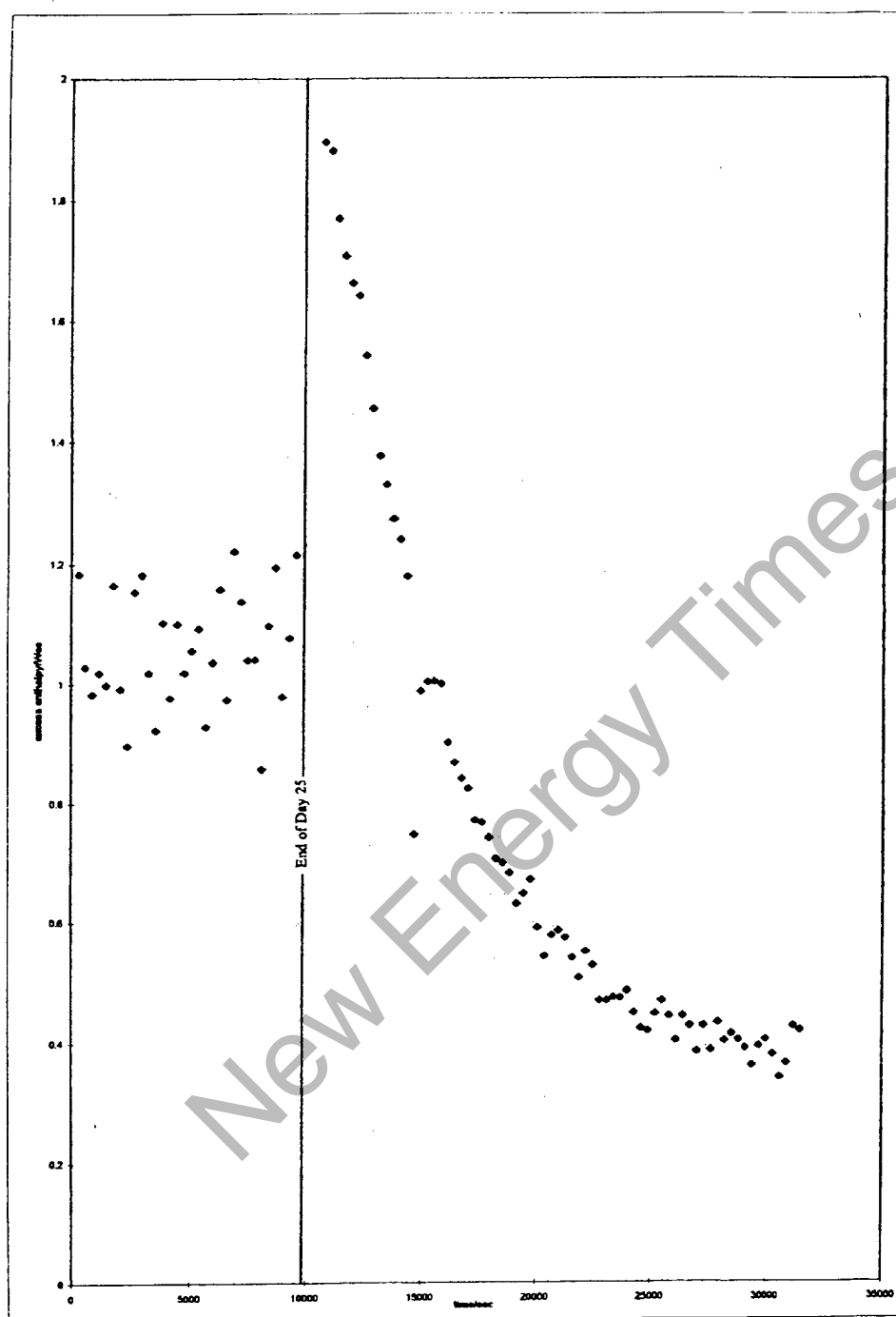


Fig. A.25 The specific rate of excess enthalpy generation/ W cm^{-3} for the last part of operation on Day 25 and the first part of operation on Day 26 of this experiment. Evaluation according to the ICARUS-Systems methodology. [Due to correction for electrode dimensions, the values for W/cm^3 should be multiplied by a factor 0.54].

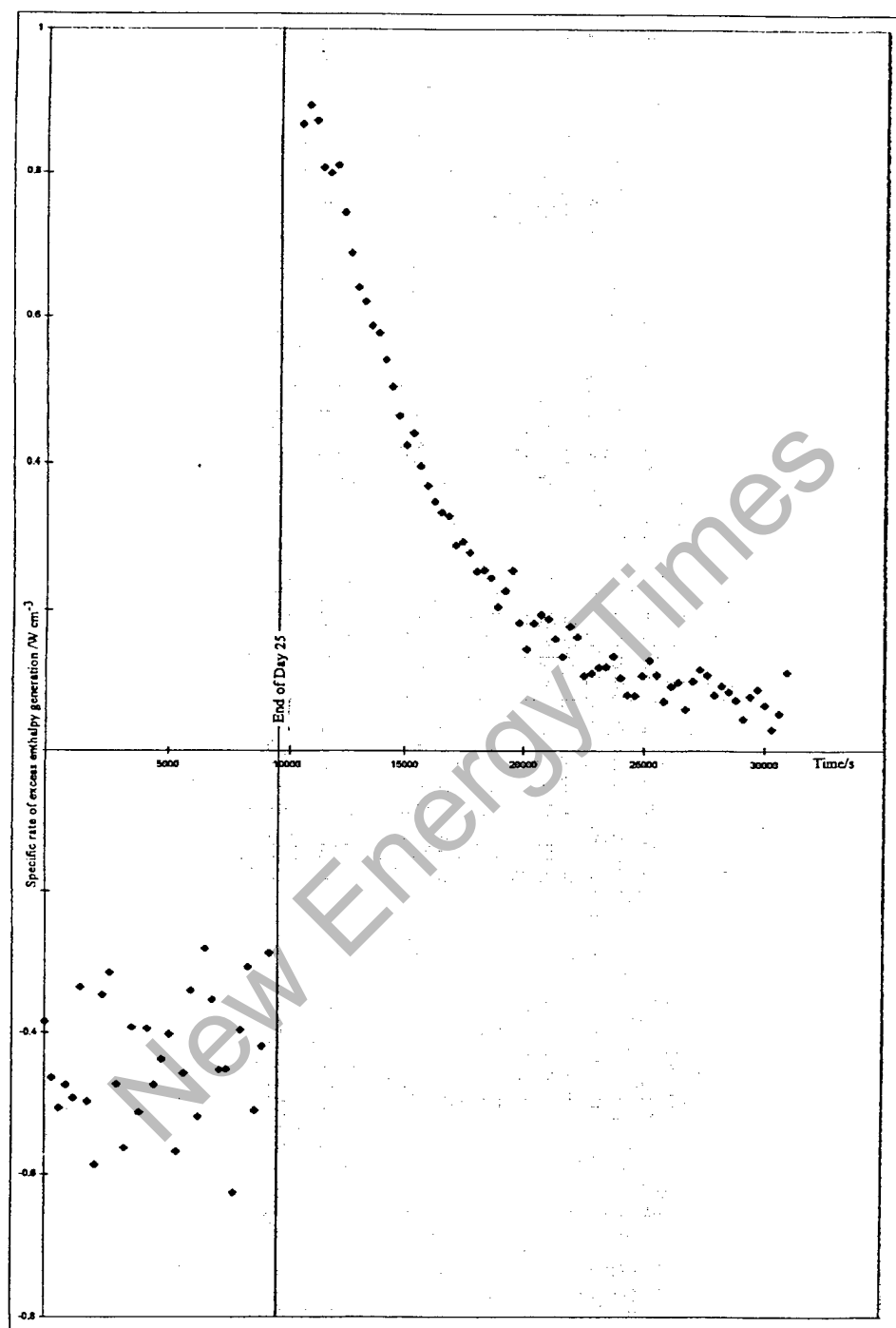


Fig. A.26 The specific rate of excess enthalpy generation for the last part of operation on Day 25 and the first part of operation on Day 26 of this experiment. Evaluation given by the group at the NHE Laboratories. [Due to correction for electrode dimensions, the values for W/cm^3 should be multiplied by a factor 0.54].

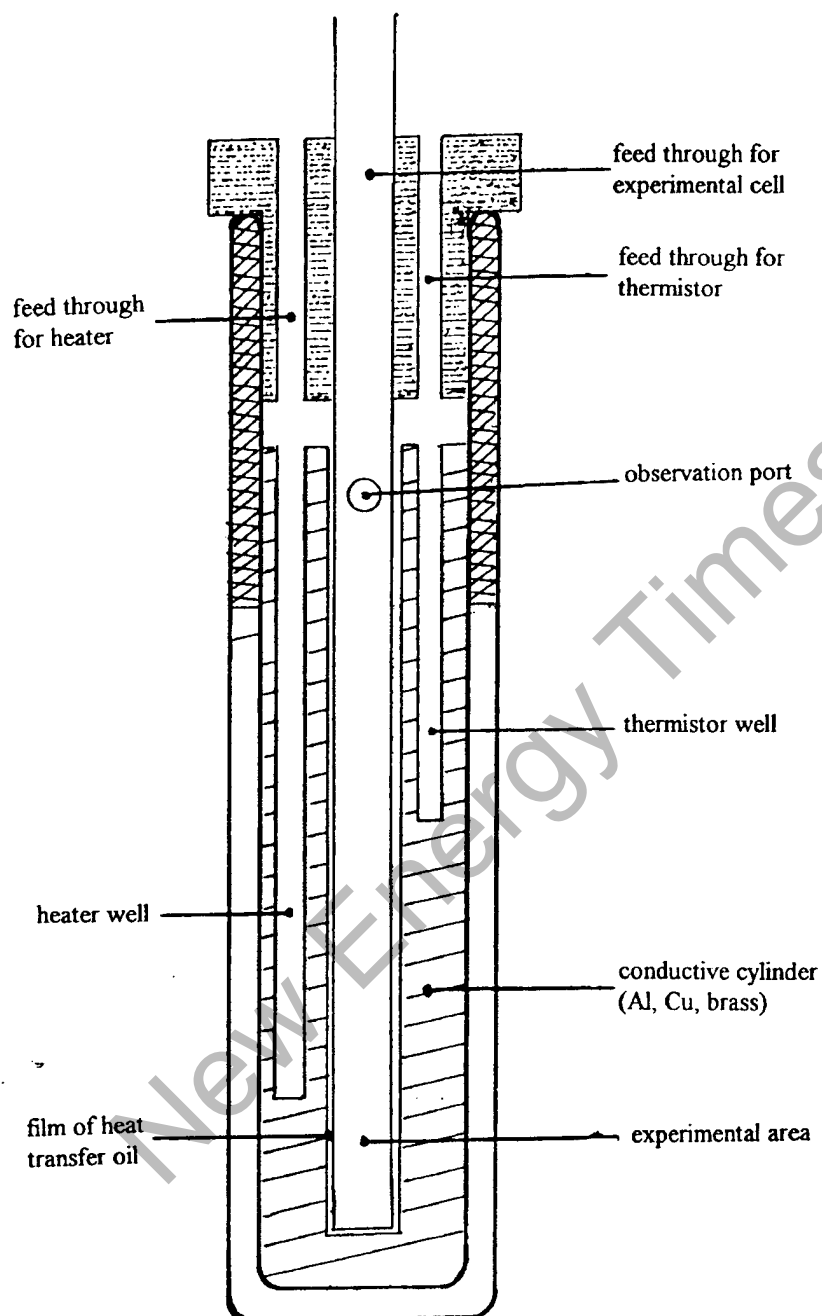


Fig. A.27 Schematic diagram of the ICARUS-14 Calorimeter. This calorimeter was originally designated as the ICARUS-4 Design.

Table A.1 The general operating characteristics of Experiment M7c2 (Pd-B)

Column 1	Time/day
Column 2	Current density/A cm ⁻²
Column 3	The maximum cell temperature for each day of operation/K
Column 4	The volume of D ₂ O added/ml
Column 5	The total volume of D ₂ O added/ml
Column 6	The volume of D ₂ O electrolysed/ml
Column 7	The total volume of D ₂ O electrolysed/ml
Column 8	The change in the volume of electrolyte at the start of the relevant day
Column 9	The change in the volume of the electrolyte at the end of the relevant day
Column 10	The total excess enthalpy generated each day/J based on the "true heat transfer coefficient, $(k_R')_{12} = 0.79350 \times 10^{-9} \text{ WK}^{-4}$ "
Column 11	The total excess enthalpy generated each day/J based on the "true heat transfer coefficient, $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$ "

Table A.1

1	2	3	4	5	6	7	8	9	10	11
1	-0.0521	300.7								
2	-0.0521	300.4			-1.22	-1.22	0	-1.22	493	3,250
3	-0.0521	303.1			-1.22	-2.44		-2.44	-1,450	1,260
4	-0.0521	303.1	4		-1.22	-3.66		-3.66	-1,420	1,580
5	-0.0521	303.1	4	4	-1.22	-4.88	0.34	-0.88	580	3,640
6	-0.0521	303.2			-1.22	-6.1		-2.1	972	4,030
7	-0.0521	303.2			-1.22	-7.32		-3.32	1,540	4,600
8	-0.0521	303.3	6		-1.22	-8.54		-4.54	1,860	4,920
9	-0.0521	303.2		10	-1.22	-9.76	1.46	0.24	-374	2,720
10	-0.0521	303.2			-1.22	-10.98		-0.98	-374	2,370
11	-0.1075	311.6	4.5		-1.22	-12.2		-2.2	-635	2,900
12	-0.1392	318	3	14.5	-2.52	-14.71	2.3	-0.21	-1,860	6,080
13	-0.1741	324.3	8	17.5	-3.26	-17.97	2.79	-0.47	-2,850	9,090
14	-0.2093	331.4	6		-4.07	-22.04		-4.54	-3,300	12,800
15	-0.2093	332.1	8	25.5	-4.9	-26.94	3.46	-1.44	6,370	27,400
16	-0.1741	325.7	6	31.5	-4.9	-31.84	4.56	-0.34	-9,770	12,400
17	-0.1741	325.2	4	35.5	-4.07	-35.91	3.66	-0.41	4,680	21,640
18	-0.1741	325.6	8		-4.07	-39.99		-4.48	6,680	23,400
19	-0.1741	325.3		43.5	-4.07	-44.06	3.52	-0.56	4,840	21,800
20	-0.1741	325.3	8		-4.07	-48.14		-4.63	6,740	23,500
21	-0.1741	325.4		51.5	-4.07	-52.21	3.37	-0.7	4,750	21,800
22	-0.1392	319.3	9	60.5	-4.07	-56.29	4.23	-4.77	6,400	23,200
23	-0.1392	319			-3.26	-59.54		0.97	3,780	16,600
24	-0.1392	318.8			-3.26	-62.8		-2.3	4,490	17,100
25	-0.2266	336.2	10	70.5	-3.26	-66.06		-5.56	5,210	17,600
26	-0.0696	306.8	8	78.5	-5.3	-71.37	4.44	-0.86	-8,720	15,900
27	-0.0696	306.7			-1.63	-72.99	7.14	5.51	1,930	7,120
28	-0.0696	306.7			-1.63	-74.62		3.88	782	5,900
29	-0.0696	306.7			-1.63	-76.25		2.25	926	6,000
30	-0.0696	306.6			-1.63	-77.88		0.62	1,090	6,140
					-1.63	-79.51		-1.01	1,310	6,340

31	-0.0696	306.6			-1.63	-81.14		-2.64	1,540	6,540
32	-0.1737	326.2	5	83.5	-4.06	-85.21	2.36	-1.7	-7,000	10,350
33	-0.1737	326.7	8	91.5	-4.06	-89.27	6.3	2.24	-9,230	8,400
34	-0.1737	326.4			-4.06	-93.34		-1.82	-6,140	11,400
35	-0.1908	329.8	4	95.5	-4.47	-97.8	2.16	-2.3	-9,600	10,300
36	-0.1388	320.2	9	104.5	-3.25	-101.05	6.7	3.45	-1,570	11,800
37	-0.1388	320			-3.25	-104.3		0.2	-639	12,600
38	-0.1388	319.7			-3.25	-107.55		-3.05	255	13,300
39	-0.1908	330.8	8	112.5	-4.47	-112.01	4.95	0.48	-6,260	14,400
40	-0.1564	323.7			-3.66	-115.67		-3.18	-428	15,200
41	-0.1735	328	9	121.5	-4.06	-119.73	5.83	1.77	-3,940	14,700
42	-0.1735	327.8			-4.06	-123.79		-2.29	-2,210	16,300
43	-0.1735	329	12	133.5	-4.06	-127.85	9.71	5.65	-12,900	6,520
44	-0.1735	329.1			-4.06	-131.91		1.59	-7,040	12,400
45	-0.1735	329			-4.06	-135.98		-2.47	-6,410	13,000
46	-0.2085	337.4	9	142.5	-4.88	-140.85	6.52	1.64	5,720	31,200
47	-0.2085	337.6			-4.88	-145.73		-3.24	9,670	35,300
48	-0.1909	335.5	10	152.5	-4.47	-150.2	6.77	2.3	6,510	30,600
49	-0.1909	336.1			-4.47	-154.67		-2.17	9,900	34,500
50	-0.1735	335.1	13	165.5	-4.06	-158.73	10.83	6.77	5,110	28,800
51	-0.1735	336.6			-4.06	-162.79		2.71	8,830	33,800
52	-0.1735	339.2			-4.06	-166.85		-1.35	12,300	39,100
53	-0.208	327.1	9	174.5	-4.87	-171.71	7.65	2.78	4,370	22,500
54	-0.208	323.9			-4.87	-176.58		-2.09	5,550	21,900
55	-0.2772	334.2	10	184.5	-6.49	-183.07	7.92	1.43	2,430	25,600
56	-0.2425	329.4	5	189.5	-5.67	-188.75	6.43	0.76	4,440	24,100
57	-0.1737	319.7	12	201.5	-4.06	-192.81	12.75	8.69	2,450	15,500
58	-0.1737	319.5			-4.06	-196.87		4.63	3,580	16,500
59	-0.1737	319.2			-4.06	-200.94		0.57	4,610	17,400
60	-0.3118	340.1	9	210.5	-7.3	-208.23	9.56	2.26	-14,600	12,800
61	-0.3484	345.4	8	218.5	-8.16	-216.39	10.27	2.11	-32,600	-436
62	-0.3484	346.2	8	226.5	-8.16	-224.55	10.11	1.95	-15,600	17,200
63	-0.3484	349.2	5	231.5	-8.16	-232.71	6.95	-1.21	-5,700	29,200
64	-0.3484	341.7			-8.16	-240.87		-9.37	-10,800	22,000
65	-0.2788	342.2			-6.53	-247.4		-15.9	-3,860	25,300

66	-0.2788	342.8	13	244.5	-6.53	-253.92	-2.9	-9.43	888	30,500
67	-0.3485	360.3			-8.16	-262.08		-17.59	-4,140	41,000
68										
69										

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Table A.2 The "raw data for the third measurement cycle of experiment M7c2 and the derived "lower bound heat transfer coefficient, $(k_R')_{11}$ " and rates of excess enthalpy production, Q , as given by the analysis provided by the group at the N.H.E. Laboratory. Values of Q have been derived using the "true heat transfer coefficient, $(k_R')_{12}$ " ($0.793504 \times 10^{-9} \text{ WK}^{-4}$) quoted by this group.

The Table also lists the averages \bar{Q} and $\bar{\bar{Q}}$ as well as $10^9(\bar{k}_R')_{11}$ and $10^9(\bar{\bar{k}}_R')_{11}$ (and the associated statistics) for the evaluations based on measurements with the short thermistor.

Columns 15 - 17 give the evaluation of $10^9(k_R')_{11}$, $10^9(\bar{k}_R')_{11}$ and $10^9(\bar{\bar{k}}_R')_{11}$ but with the inclusion of the enthalpy input $\Delta Q = 0.2500\text{W}$ due to the calibration heater.

Column 1	The elapsed time/s
Column 2	The temperature of the water thermostat/K
Column 3	The temperature measured with the "short thermistor"/K
Column 4	The temperature measured with the "long thermistor"/K
Column 5	The Cell Voltage/V
Column 6	The Cell Current/A
Column 7	$10^9(k_R')_{11}/\text{WK}^{-4}$ for measurements in Column 3
Column 8	Q/W for measurements in Column 3
Column 9	$10^9(\bar{k}_R')_{11}/\text{WK}^{-4}$ for measurements in Column 4
Column 10	Q/W for measurements in Column 4
Column 11	\bar{Q}/W of the values in Column 8
Column 12	$\bar{\bar{Q}}/W$ of the values in Column 11
Column 13	$10^9(\bar{k}_R')_{11}/\text{WK}^{-4}$ σ/WK^{-4} and $\sigma/(\bar{k}_R')_{11}/\%$ of the values in Column 9
Column 14	$10^9(\bar{\bar{k}}_R')_{11}/\text{WK}^{-4}$ σ/WK^{-4} and $\sigma/(\bar{\bar{k}}_R')_{11}/\%$ of the values in Column 13

- Column 15 $10^9(k_R')_{11}/WK^{-4}$ for measurements in Column 3 but with inclusion of the enthalpy input $\Delta Q = 0.2500W$ due to the calibration heater.
- Column 16 $10^9(\overline{k_R'})_{11}/WK^{-4}$ σ/WK^{-4} and $\sigma/(\overline{k_R'})_{11}/\%$ of the values in Column 15
- Column 17 $10^9(\overline{\overline{k_R'}})_{11}/WK^{-4}$ σ/WK^{-4} and $\sigma/(\overline{\overline{k_R'}})_{11}/\%$ of the values in Column 16

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Table A.2

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
300	295.151	300.353	300.349	4.5904	0.15035	0.840812	-0.0258	0.839983	-0.0253							
600	295.203	300.354	300.349	4.5965	0.15035	0.844238	-0.0276	0.840573	-0.0256							
900	295.197	300.351	300.35	4.5864	0.15035	0.837996	-0.0242	0.838163	-0.0243							
1200	295.191	300.354	300.349	4.589	0.15035	0.834268	-0.0222	0.839597	-0.0251							
1500	295.206	300.353	300.349	4.5912	0.15035	0.838914	-0.0247	0.838081	-0.0242							
1800	295.196	300.355	300.351	4.5895	0.15035	0.838025	-0.0243	0.841693	-0.0262							
2100	295.204	300.353	300.347	4.5933	0.15035	0.846685	-0.0289	0.841686	-0.0262							
2400	295.213	300.351	300.351	4.5878	0.15038	0.840981	-0.0258	0.835468	-0.0233							
2700	295.217	300.353	300.35	4.595	0.15035	0.840212	-0.0253	0.849754	-0.0305							
3000	295.21	300.353	300.347	4.595	0.15035	0.845111	-0.028	0.849135	-0.0302							
3300	295.199	300.351	300.346	4.5857	0.15035	0.845623	-0.0284	0.840458	-0.0255							
3600	295.2	300.348	300.346	4.5845	0.15035	0.839948	-0.0238	0.840285	-0.0254							
3900	295.2	300.352	300.345	4.5821	0.15035	0.834114	-0.0221	0.838286	-0.0243							
4200	295.202	300.35	300.346	4.5787	0.15036	0.83538	-0.0228	0.828532	-0.019							
4500	295.19	300.353	300.341	4.5759	0.15036	0.83321	-0.0217	0.833542	-0.0218							
4800	295.204	300.351	300.347	4.5787	0.15038	0.840764	-0.0257	0.848853	-0.0301							
5100	295.201	300.35	300.343	4.5734	0.15035	0.838206	-0.0243	0.837875	-0.0241							
5400	295.203	300.349	300.346	4.5794	0.15036	0.838904	-0.0247	0.8384	-0.0233							
5700	295.209	300.349	300.344	4.5745	0.15036	0.835492	-0.0228	0.843858	-0.0273							
6000	295.187	300.35	300.342	4.5771	0.15035	0.835515	-0.0229	0.838847	-0.0236							
6300	295.195	300.348	300.343	4.5743	0.15035	0.840838	-0.0258	0.832667	-0.0213							
6600	295.197	300.346	300.344	4.5784	0.15035	0.835118	-0.0228	0.836954	-0.0236							
6900	295.199	300.349	300.343	4.5837	0.15036	0.833452	-0.0217	0.83496	-0.0247							
7200	295.195	300.349	300.344	4.579	0.15035	0.837468	-0.0239	0.838305	-0.0244							
7500	295.197	300.348	300.342	4.5718	0.15036	0.837548	-0.024	0.838551	-0.0245							
7800	295.191	300.347	300.342	4.5794	0.15036	0.837336	-0.0239	0.838171	-0.0243							
8100	295.198	300.347	300.341	4.5763	0.15036	0.830237	-0.02	0.835743	-0.0229							
8400	295.184	300.351	300.343	4.5806	0.15035	0.83592	-0.0231	0.831247	-0.0205							
8700	295.211	300.347	300.345	4.5749	0.15036	0.843781	-0.0273	0.838095	-0.0242							
9000	295.204	300.347	300.343	4.5639	0.15036	0.830597	-0.0201	0.83728	-0.0238							
9300	295.207	300.348	300.343	4.5674	0.15036	0.837666	-0.024	0.836671	-0.0245							
9600	295.198	300.345	300.341	4.5674	0.15035	0.832401	-0.0211	0.837576	-0.0234							
9900	295.21	300.35	300.341	4.5659	0.15035	0.839063	-0.0247	0.840574	-0.0255							
10200	295.211	300.342	300.338	4.565	0.15035	0.843332	-0.027	0.833449	-0.0218							
10500	295.207	300.345	300.343	4.5701	0.15035	0.830055	-0.0198	0.830387	-0.02							
10800	295.21	300.346	300.342	4.5679	0.15035	0.834268	-0.0221	0.839455	-0.0248							
11100	295.196	300.348	300.341	4.5678	0.15034	0.830501	-0.0201	0.840348	-0.0255							
11400	295.195	300.348	300.338	4.5672	0.15035	0.841879	-0.0263	0.83905	-0.0247							
11700	295.218	300.34	300.338	4.5631	0.15034	0.83968	-0.025	0.833976	-0.0219							
12000	295.204	300.346	300.34	4.5702	0.15035	0.827941	-0.0187	0.833451	-0.0217							
12300	295.206	300.345	300.34	4.5689	0.15035	0.835563	-0.0228	0.836419	-0.0233							
12600	295.214	300.348	300.34	4.5824	0.15035	0.837895	-0.0241	0.838902	-0.0248							
12900	295.207	300.343	300.338	4.573	0.15035	0.843234	-0.027	0.83805	-0.0241							

Table A.3 Abridged $(k_R')_{11}$ spreadsheet for the third measurement cycle of experiment M7c2. The evaluation shown in Columns 2 - 8 has included all the corrections to that given by the group in the N.H.E. Laboratories with the exception of the use of $C_{pM} = 490 \text{ JK}^{-1}$. The evaluation in Columns 9 - 12 includes the correction of this parameter to $C_{pM} = 450 \text{ JK}^{-1}$

Column 1	The elapsed time/s
Column 2	The total input power/W
Column 3	Values of $-C_{pM}(d\theta/dt)/f(\theta) \text{ /WK}^{-4}$
Column 4	Values of input power/ $(\theta) \text{ /WK}^{-4}$
Column 5	The derived values of $10^9(k_R')_{161}$, $10^9(k_R')_{171}/\text{WK}^{-4}$, C_{pM}/JK^{-1} and the relevant regression coefficients. The arrows indicate the ranges of the fitting procedures
Column 6	The derived values of $10^9(k_R')_{181}/\text{WK}^{-4}$, C_{pM}/JK^{-1} and the relevant regression coefficients. The arrows indicate the ranges of the fitting procedures; the dotted line indicates the linkage of the data for measurements in $t_1 < t < t_2$ with those for $t_2 < T$.
Column 7	$10^9(k_R')_{11}$ derived from Columns 3 and 4
Column 8	The 11-point running mean of $10^9(k_R')_{11}$
Column 9	Values of $-450(d\theta/dt)/f(\theta) \text{ /WK}^{-4}$
Column 10	Values of $10^9(k_R')_{11} \text{ /WK}^{-4}$ based on Columns 4 and 9.
Column 11	The 11-point running mean /WK^{-4} of $10^9(k_R')_{11}$ shown in Column 10
Column 12	The rate of excess enthalpy generation/W based on the "true heat transfer coefficient, $(k_R')_{12}'' = 0.85065 \times 10^{-9} \text{ WK}^{-4}$

Table A.3

[illegible]

9000	0.45528	0.00301	0.83826			0.84127	0.84004	0.00276	0.84102	0.83944	0.0013
9300	0.45528	-0.0015	0.8375			0.836	0.83942	-0.00138	0.83612	0.83886	0.00396
9600	0.45506	0.00451	0.83815			0.84266	0.84047	0.00414	0.84229	0.8398	0.0006
9900	0.45492	0.00754	0.8394			0.84708	0.84082	0.00692	0.84632	0.84013	0.00177
10200	0.45569	-0.00602	0.83968			0.83366	0.83938	-0.00552	0.83416	0.83879	0.00501
10500	0.45536	-0.00151	0.83938			0.83787	0.83928	-0.00138	0.838	0.83922	0.00292
10800	0.45533	-0.003	0.8371			0.8341	0.83931	-0.00276	0.83435	0.83924	0.00491
11100	0.45525	0.009	0.83647			0.84547	0.8403	0.00827	0.84474	0.84017	-0.00075
11400	0.4546	0.00302	0.84027			0.84329	0.83991	0.00277	0.84304	0.83982	0.00017
11700	0.4557	-0.00752	0.83906			0.83154	0.83904	-0.00689	0.83117	0.83904	0.00608
12000	0.45551	0	0.83918			0.83918	0.83971	0	0.83918	0.83963	0.00226
12300	0.45453	0.00301	0.83849			0.8415	0.83962	0.00276	0.84125	0.83954	0.00114
12600	0.45612	0.00602	0.84081			0.84683	0.83996	0.00552	0.84633	0.83984	-0.00162
12900	0.45587	-0.0015	0.83987			0.83837	0.83991	-0.00138	0.83849	0.83981	0.00263
13200	0.45521	-0.00151	0.83912			0.83761	0.83977	-0.00139	0.83773	0.83968	0.00198
13500	0.45491	0.00301	0.83793			0.84094	0.84019	0.00276	0.84069	0.84006	0.00143
13800	0.4548	0	0.83696			0.83696	0.8398	0	0.83696	0.8397	0.00345
14100	0.45427	0.0015	0.8363			0.8378	0.83992	0.00138	0.83768	0.8398	0.00306
14400	0.45515	0.00602	0.83887			0.84489	0.83962	0.00552	0.84439	0.83952	-0.00059
14700	0.45545	0.00301	0.83881			0.84182	0.83942	0.00276	0.84157	0.83934	0.00094
15000	0.45528	-0.00301	0.83914			0.83613	0.83962	-0.00276	0.83639	0.83955	0.00375
15300	0.45489	-0.00301	0.83793			0.83492	0.8401	-0.00276	0.83517	0.84001	0.00178
15600	0.45552	0.00451	0.83829			0.8428	0.8396	0.00414	0.84243	0.83954	0.00046
15900	0.45566	0.00301	0.84048			0.84349	0.83948	0.00276	0.84324	0.83943	0.00002
16200	0.45474	-0.00453	0.84069			0.83616	0.83921	-0.00416	0.83653	0.83919	0.00365
16500	0.45429	0.00301	0.83682			0.83983	0.83841	0.00276	0.83958	0.83842	0.002
16800	0.45497	0.00603	0.84017			0.8462	0.83866	0.00554	0.84571	0.83863	-0.00133
17100	0.45406	-0.00451	0.83596			0.83145	0.83923	-0.00414	0.83182	0.83937	0.00621
17400	0.45405	0	0.83654			0.83654	0.83931	0	0.83654	0.83944	0.00364
17700	0.4542	0.00302	0.83891			0.84193	0.83875	0.00277	0.84168	0.83891	0.00084
18000	0.45394	-0.0015	0.83447			0.83297	0.83885	-0.00138	0.83309	0.83898	0.00552
18300	0.45431	0.00151	0.83735			0.83886	0.83901	0.00139	0.83874	0.83916	0.00244
18600	0.45467	0.00752	0.83675			0.84127	0.83932	0.00691	0.84366	0.83934	-0.00024
18900	0.45515	0.00452	0.8391			0.84362	0.84059	0.00415	0.84325	0.84056	-0.00002
19200	0.45518	-0.00151	0.83883			0.83732	0.84041	-0.00139	0.83744	0.84041	0.00313

19500	0.45495	0	0.8373				0.8373	0.84015	0	0.8373	0.84018	0.00321
19800	0.45525	0.00151	0.84008				0.84159	0.84041	0.00139	0.84147	0.84047	0.00094
20100	0.45429	0.01055	0.838				0.84855	0.84115	0.00969	0.84769	0.84119	-0.00244
20400	0.4544	0.00754	0.8384				0.84594	0.84158	0.00692	0.84532	0.84134	-0.00116
20700	0.45416	-0.00301	0.83763				0.83462	0.84036	-0.00276	0.83487	0.84022	0.00451
21000	0.45524	-0.00151	0.84058				0.83907	0.83995	-0.00139	0.83919	0.8399	0.00216
21300	0.45588	-0.00452	0.84033				0.83581	0.84069	-0.00415	0.83618	0.84044	0.00379
21600	0.45647	0.00302	0.84395				0.84697	0.84087	0.00277	0.84672	0.84084	-0.00192
21900	0.45449	0.00902	0.83698				0.846	0.8402	0.00828	0.84526	0.84024	-0.00114
22200	0.45527	-0.00903	0.83922				0.83019	0.83886	-0.00829	0.83093	0.839	0.00663
22500	0.45497	-0.00603	0.8394				0.83337	0.83956	-0.00554	0.83386	0.83963	0.00503
22800	0.45495	0.00602	0.83765				0.84543	0.84033	0.00553	0.84318	0.84032	-0.00002
23100	0.45455	0.00754	0.83898				0.84367	0.8407	0.00692	0.8459	0.84064	-0.0015
23400	0.45497	0	0.84107				0.84107	0.83975	0	0.84107	0.83975	0.00111
23700	0.45482	-0.00601	0.83725				0.83124	0.83846	-0.00552	0.83173	0.83857	0.00619
24000	0.45498	0.00601	0.83629				0.8423	0.83915	0.00552	0.84181	0.8392	0.00071
24300	0.45408	0.01054	0.83704				0.84758	0.84183	0.00968	0.84672	0.84176	-0.00196
24600	0.45504	0.00151	0.83836				0.83987	0.84103	0.00139	0.83975	0.84117	0.00182
24900	0.4561	-0.00452	0.84108				0.83656	0.84017	-0.00415	0.83693	0.84014	0.00335
25200	0.45398	-0.00603	0.8378				0.83177	0.84037	-0.00554	0.83226	0.84031	0.00587
25500	0.45459	0	0.8378				0.8378	0.84108	0	0.8378	0.84105	0.00287
25800	0.46166	0.00906	0.85376				0.86282	0.84106	0.00832	0.86208	0.84107	-0.01027
26100	0.45438	-0.00151	0.8381				0.83659	0.84042	-0.00139	0.83671	0.84048	0.00345
26400	0.45398	-0.00301	0.83732				0.83431	0.84039	-0.00276	0.83456	0.84045	0.00461
26700	0.45414	0.00452	0.83875				0.84327	0.84009	0.00415	0.8429	0.84014	0.00009
27000	0.45461	0.00151	0.83851				0.83902	0.8407	0.00139	0.8399	0.84069	0.00171
27300	0.45444	0.00151	0.8406				0.84211	0.84117	0.00139	0.84199	0.84074	0.00057
27600	0.45404	0.00301	0.83746				0.84047	0.83452	0.00276	0.84022	0.83897	0.00153
27900	0.45425	0.00151	0.83802				0.83953	0.84044	0.00139	0.83941	0.84011	0.00197
28200	0.45383	-0.00301	0.83629				0.83328	0.84097	-0.00276	0.83353	0.84117	0.00516
28500	0.45393	0.00151	0.83694				0.83845	0.83999	0.00139	0.83833	0.84091	0.00255
28800	0.45398	0.00453	0.83848				0.84301	0.84022	0.00416	0.84264	0.84027	0.00021
29100	0.45468	0.00603	0.83867				0.8447	0.84055	0.00554	0.84421	0.84022	-0.00065
29400	0.45468	0.00604	0.84062				0.84666	0.84009	0.00555	0.84617	0.84069	-0.00171
29700	0.45416	0	0.84013				0.84013	0.84072	0	0.84013	0.8406	0.00155

30000	0.45402	-0.00452	0.83701				0.83249	0.84174	-0.00415	0.83286	0.84156	0.0055
30300	0.45438	0.00301	0.8386				0.84161	0.83867	0.00276	0.84136	0.84184	0.00089
30600	0.45459	0.00301	0.84271				0.84572	0.83529	0.00276	0.84547	0.84155	-0.00134
30900	0.4557	-0.00603	0.84138				0.83535	0.83189	-0.00554	0.83584	0.84098	0.00387
31200	0.45399	0.00605	0.84044				0.84649	0.83009	0.00556	0.845	0.84169	-0.00163
31500	0.45413	0.00453	0.83994				0.84447	0.82776	0.00416	0.8441	0.84173	-0.00061
31800	0.70356	-0.00226	1.2779				1.27519		-0.02076	1.27704	0.84154	
32100	0.7051	-0.26179	1.29898				1.03719		-0.24042	1.05856	0.84204	
32400	0.70321	-0.45041	1.25513				0.80472	0.82535	-0.41364	0.84149	0.84154	0.00082
32700	0.70179	-0.41048	1.21637				0.80589	0.82382	-0.37697	0.8394	0.84204	0.00205
33000	0.70112	-0.37648	1.18372				0.80724	0.82116	-0.34575	0.83797	0.84124	0.00294
33300	0.70187	-0.3316	1.15849				0.82689	0.81976	-0.30453	0.85396	0.84147	-0.00668
33600	0.70056	-0.31997	1.1344				0.81443	0.81584	-0.29385	0.84055	0.83926	0.00147
33900	0.70214	-0.30325	1.10928				0.80603	0.81205	-0.2785	0.83079	0.83711	0.00768
34200	0.70519	-0.27013	1.09491				0.82478	0.81299	-0.24808	0.84683	0.83607	-0.00252
34500	0.69713	-0.24814	1.06459				0.81645	0.81213	-0.22788	0.83671	0.83508	0.00406
34800	0.69704	-0.2253	1.04524				0.81994	0.81236	-0.20691	0.83833	0.83373	0.00305
35100	0.69542	-0.22456	1.02792		0.83808		0.80336	0.81236	-0.20623	0.82169	0.83126	0.01435
35400	0.69547	-0.20933	1.01268		454		0.80285	0.81293	-0.19224	0.82044	0.82974	0.01542
35700	0.69474	-0.18442	0.99943		0.9956		0.82524	0.81113	-0.16937	0.83007	0.8286	0.00891
36000	0.69373	-0.17319	0.98756				0.9995	0.81437	-0.15905	0.82851	0.826	0.0101
36300	0.69334	-0.16401	0.97375				0.80974	0.81033	-0.15062	0.82313	0.82535	0.01406
36600	0.6948	-0.15091	0.96534				0.81443	0.81192	-0.13859	0.82675	0.82478	0.0116
36900	0.69238	-0.14183	0.95415				0.81232	0.81259	-0.13025	0.8239	0.8245	0.01376
37200	0.69336	-0.1393	0.94618				0.80688	0.81177	-0.12793	0.81825	0.82369	0.01803
37500	0.69225	-0.12804	0.9358				0.80776	0.81132	-0.11759	0.81821	0.82246	0.01823
37800	0.69961	-0.11924	0.93715				0.81791	0.81099	-0.09763	0.83952	0.82149	0.00248
38100	0.69799	-0.10631	0.9272				0.82089	0.81053	-0.09515	0.83204	0.82043	0.00813
38400	0.69142	-0.10362	0.91381				0.81019	0.80972	-0.09516	0.81865	0.81908	0.0183
38700	0.68971	-0.10299	0.90603	0.81319			0.8218	0.80304	-0.09458	0.81145	0.81768	0.02389
39000	0.68936	-0.08826	0.89765	488			472	0.80939	-0.08106	0.8166	0.81655	0.02014
39300	0.69164	-0.08693	0.89762	0.9977			0.9993	0.81069	-0.07983	0.81779	0.81632	0.01928
39600	0.68947	-0.08308	0.88778					0.8047	-0.0763	0.81148	0.81609	0.02433
39900	0.68844	-0.07751	0.8831					0.80559	-0.07118	0.81192	0.81377	0.02408
40200	0.68772	-0.06965	0.87536				0.80571	0.80618	-0.06396	0.8114	0.81143	0.02467

40500	0.68779	-0.06629	0.87234						0.80605	0.80611	-0.06088	0.81146	0.81099	0.02471
40800	0.68872	-0.06099	0.87173						0.81074	0.80583	-0.05601	0.81572	0.81041	0.02138
41100	0.68799	-0.05447	0.86565						0.81118	0.80578	-0.05002	0.81563	0.80999	0.02158
41400	0.68725	-0.05934	0.86101						0.80167	0.80622	-0.0545	0.80651	0.81009	0.02895
41700	0.68751	-0.05712	0.85875						0.80163	0.80559	-0.05246	0.80629	0.80924	0.02921
42000	0.68729	-0.05392	0.85616	✓	0.80784	✓	0.81801		0.80224	0.80502	-0.04952	0.80664	0.80846	0.029
42300	0.68822	-0.04656	0.85294		497		475		0.80638	0.80526	-0.04276	0.81018	0.80845	0.02629
42600	0.68693	-0.03833	0.84846		0.9984		0.9991		0.81013	0.80475	-0.0352	0.81326	0.80772	0.02387
42900	0.68691	-0.0383	0.84775						0.80945	0.80456	-0.03517	0.81258	0.8073	0.02444
43200	0.68713	-0.0462	0.84496						0.79876	0.80487	-0.04243	0.80253	0.80739	0.0327
43500	0.68665	-0.04203	0.84137						0.79934	0.80477	-0.0386	0.80277	0.80711	0.03262
43800	0.6871	-0.03198	0.84073						0.80875	0.80518	-0.02937	0.81136	0.80728	0.02564
44100	0.68568	-0.03191	0.83707						0.80516	0.80501	-0.02931	0.80777	0.80693	0.02863
44400	0.68611	-0.02584	0.83484						0.809	0.80454	-0.02173	0.81311	0.80636	0.02433
44700	0.68593	-0.02879	0.83391						0.80512	0.80394	-0.02644	0.80747	0.80563	0.02899
45000	0.68509	-0.03174	0.83231						0.80057	0.80429	-0.02915	0.80316	0.8058	0.03255
45300	0.68584	-0.02271	0.82938	✓	0.80633				0.80667	0.80461	-0.02086	0.80852	0.80596	0.02826
45600	0.68563	-0.02266	0.82719		499				0.80453	0.80439	-0.02081	0.80638	0.80563	0.0301
45900	0.68633	-0.02367	0.82855		0.9987				0.80498	0.80483	-0.02174	0.80691	0.80593	0.02963
46200	0.68466	-0.02164	0.8245						0.80286	0.80453	-0.01987	0.80463	0.80554	0.0316
46500	0.68444	-0.02163	0.82423						0.8026	0.80414	-0.01986	0.80437	0.80508	0.03122
46800	0.68546	-0.0206	0.82344						0.80284	0.8046	-0.01892	0.80452	0.8054	0.03176
47100	0.68567	-0.01767	0.82401						0.80634	0.80432	-0.01623	0.80778	0.80505	0.02903
47400	0.68554	-0.01274	0.82278						0.81004	0.80407	-0.0117	0.81108	0.80474	0.02631
47700	0.68511	-0.01368	0.81938						0.8057	0.80382	-0.01256	0.80682	0.80441	0.02996
48000	0.68596	-0.01953	0.82033						0.8008	0.80351	-0.01794	0.80239	0.80403	0.03366
48300	0.685	-0.01169	0.81738						0.80569	0.80385	-0.01074	0.80664	0.80427	0.03017
48600	0.68508	-0.01463	0.81817						0.80354	0.80328	-0.01344	0.80473	0.80366	0.03174
48900	0.68428	-0.01362	0.8154						0.80178	0.80307	-0.01251	0.80289	0.8034	0.03335
49200	0.68452	-0.01264	0.81488						0.80224	0.80332	-0.01161	0.80327	0.80356	0.03306
49500	0.68443	-0.01358	0.81297						0.79934	0.80302	-0.01247	0.8005	0.80322	0.03546
49800	0.68489	-0.00679	0.81322						0.80643	0.80281	-0.00624	0.80698	0.80296	0.03001
50100	0.68515	-0.01647	0.81298						0.79651	0.80237	-0.01513	0.79785	0.8025	0.03772
50400	0.6849	-0.0097	0.81379						0.80409	0.80178	-0.00891	0.80488	0.80193	0.03175
50700	0.685	-0.00097	0.81371						0.81274	0.8021	-0.00089	0.81282	0.80219	0.02507

51000	0.6839	-0.00872	0.8111				0.80239	0.80265	-0.00801	0.80309	0.80265	0.0333
51300	0.68465	-0.01258	0.81114				0.79856	0.80309	-0.01155	0.79959	0.80302	0.03629
51600	0.68415	-0.01065	0.8114				0.80075	0.8028	-0.00978	0.80162	0.80269	0.03454
51900	0.68527	-0.01549	0.81258				0.79709	0.80589	-0.01423	0.79835	0.80297	0.03729
52200	0.68557	-0.0058	0.81116				0.80536	0.80772	-0.00533	0.80583	0.80198	0.03105
52500	0.68373	-0.00097	0.80923				0.80826	0.80802	-0.00089	0.80834	0.80081	0.02891
52800	0.68415	-0.00386	0.8081				0.80424	0.80974	-0.00036	0.80775	0.80084	0.02946
53100	0.6836	-0.00386	0.80707				0.80321	0.81206	-0.00355	0.80353	0.80087	0.03305
53400	0.43406	0.0029	0.51324				0.51614		0.00266	0.5159	0.80077	
53700	0.43526	0.16243	0.51521				0.67764		0.14917	0.66438	0.80089	
54000	0.43444	0.30301	0.52504				0.82805	0.81376	0.27827	0.80331	0.80059	0.03245
54300	0.43601	0.28688	0.53742		0.81801		0.8243	0.81577	0.26346	0.80088	0.80063	0.03379
54600	0.43581	0.2687	0.54729		475		0.81599	0.81686	0.24677	0.79406	0.80067	0.03859
54900	0.43757	0.262	0.55937		0.9991		0.82137	0.8179	0.24061	0.79998	0.80108	0.03327
55200	0.43876	0.25298	0.57107				0.82405	0.81947	0.23233	0.8034	0.80116	0.03005
55500	0.43831	0.23977	0.57967				0.81944	0.82135	0.2202	0.79987	0.80159	0.03224
55800	0.43967	0.22829	0.59088				0.81917	0.82106	0.20965	0.80053	0.80215	0.03123
56100	0.44056	0.21618	0.60114				0.81732	0.82097	0.19853	0.79967	0.80239	0.03138
56400	0.44043	0.21021	0.60951				0.81972	0.82142	0.19305	0.80256	0.80408	0.02885
56700	0.44145	0.20265	0.61887	0.81337			0.82152	0.82129	0.18611	0.80498	0.80471	0.02675
57000	0.4418	0.19524	0.62871	476			0.82395	0.82151	0.1793	0.80801	0.80564	0.02422
57300	0.44294	0.18519	0.63965	0.9955			0.82484	0.82211	0.17007	0.80972	0.80692	0.02268
57600	0.44254	0.17757	0.64571		0.8218		0.82328	0.82288	0.16307	0.80878	0.80832	0.02308
57900	0.44294	0.16746	0.65354		472		0.821	0.82393	0.15379	0.80733	0.80995	0.02381
58200	0.44318	0.15843	0.66145		0.9993		0.81988	0.82462	0.1455	0.80695	0.81124	0.02091
58500	0.44402	0.15752	0.669				0.82652	0.82484	0.14466	0.81366	0.81209	0.0191
58800	0.44483	0.1481	0.67789				0.82599	0.82487	0.13601	0.8139	0.81273	0.01873
59100	0.44489	0.14331	0.6843				0.82761	0.82578	0.13161	0.81591	0.81414	0.01723
59400	0.44481	0.13822	0.69067				0.82889	0.82666	0.12694	0.81761	0.81552	0.01598
59700	0.44454	0.12927	0.69807				0.82734	0.82697	0.11872	0.81679	0.81637	0.01635
60000	0.44637	0.1176	0.70633				0.82393	0.82739	0.108	0.81433	0.8173	0.01775
60300	0.44582	0.11332	0.71095				0.82427	0.8279	0.10407	0.81502	0.81829	0.01717
60600	0.44686	0.11792	0.71694	0.8298			0.83486	0.82802	0.10829	0.82523	0.81887	0.0107
60900	0.44697	0.10967	0.72329	505	0.82524		0.83296	0.82796	0.10072	0.82401	0.81926	0.01136
61200	0.44691	0.09468	0.72969	0.9979	470		0.82437	0.8279	0.08695	0.81664	0.81967	0.01577

61500	0.4476	0.08984	0.73473				0.9995	0.82457	0.82783	0.08251	0.81724	0.81999	0.01532
61800	0.44775	0.093	0.73912					0.83212	0.82873	0.08541	0.82453	0.82122	0.01081
62100	0.44817	0.08517	0.74208					0.82725	0.82945	0.07822	0.8203	0.82225	0.01333
62400	0.44733	0.08287	0.74409					0.82696	0.82905	0.07611	0.8202	0.82224	0.01333
62700	0.44797	0.07673	0.75155					0.82828	0.82899	0.07047	0.82202	0.82251	0.01212
63000	0.4474	0.07416	0.75234					0.8265	0.82962	0.06811	0.82045	0.82342	0.01303
63300	0.4482	0.07596	0.75797					0.83393	0.83065	0.06976	0.82773	0.82433	0.00864
63600	0.44867	0.06941	0.76268					0.83209	0.83051	0.06374	0.82642	0.82451	0.00937
63900	0.44774	0.06686	0.76369	0.83188				0.83055	0.83112	0.0614	0.82509	0.82536	0.01011
64200	0.44872	0.0643	0.76797	510				0.83227	0.83164	0.05905	0.82702	0.82613	0.00895
64500	0.44947	0.05763	0.77366	0.9987				0.83129	0.8313	0.05293	0.82659	0.82608	0.00914
64800	0.44814	0.05652	0.77535					0.83187	0.83141	0.05191	0.82726	0.82643	0.0087
65100	0.44838	0.05111	0.77954					0.83065	0.83146	0.04694	0.82648	0.82671	0.00911
65400	0.44931	0.05121	0.78268					0.83389	0.83144	0.04703	0.82971	0.82691	0.00723
65700	0.44866	0.04987	0.78282					0.83269	0.83151	0.0458	0.82862	0.8272	0.00784
66000	0.44845	0.03865	0.78595					0.8246	0.83194	0.0355	0.82145	0.82782	0.01189
66300	0.44819	0.04018	0.78746					0.82764	0.83218	0.0369	0.82436	0.82822	0.01021
66600	0.44917	0.04453	0.79001					0.83454	0.83206	0.0409	0.83091	0.82831	0.00647
66900	0.44839	0.04036	0.79149					0.83185	0.8317	0.03707	0.82856	0.82813	0.00777
67200	0.44871	0.03756	0.79373					0.83129	0.83195	0.03449	0.82822	0.82852	0.00794
67500	0.44994	0.0391	0.79791					0.83701	0.83251	0.03591	0.83382	0.82922	0.00476
67800	0.44904	0.03488	0.79904					0.83392	0.83324	0.03203	0.83107	0.83006	0.00629
68100	0.44942	0.02912	0.80141					0.83053	0.83321	0.02674	0.82815	0.83019	0.00791
68400	0.44936	0.02619	0.8005					0.82669	0.8329	0.02405	0.82955	0.83007	0.00914
68700	0.44907	0.03219	0.80449					0.83668	0.83301	0.02956	0.83405	0.8303	0.00457
69000	0.4493	0.03226	0.80664					0.8389	0.83277	0.02963	0.83627	0.83023	0.00332
69300	0.44972	0.02351	0.80911					0.83262	0.83265	0.02159	0.8307	0.83022	0.00641
69600	0.44987	0.01764	0.80959					0.82723	0.83339	0.0162	0.82579	0.83103	0.00913
69900	0.45018	0.02059	0.81062					0.83121	0.83334	0.01891	0.82953	0.8311	0.00705
70200	0.44988	0.02208	0.81091					0.83299	0.83318	0.02028	0.83119	0.83108	0.00612
70500	0.4497	0.01623	0.81244					0.82867	0.83303	0.01491	0.82735	0.83104	0.00822
70800	0.44904	0.02363	0.81204					0.83567	0.83316	0.0217	0.83374	0.83127	0.00468
71100	0.44997	0.02665	0.81549					0.84214	0.83353	0.02447	0.83996	0.83169	0.00123
71400	0.45027	0.01187	0.81808					0.82995	0.83385	0.0109	0.82898	0.83205	0.00727
71700	0.44988	0.00742	0.81753					0.82495	0.83423	0.00681	0.82434	0.83248	0.00982

72000	0.45047	0.01632	0.81863				0.83495	0.83428	0.01499	0.83362	0.8326	0.00471
72300	0.44996	0.02083	0.81959				0.84042	0.8348	0.01913	0.83872	0.83315	0.0019
72600	0.45014	0.01491	0.82168				0.83659	0.83482	0.01369	0.83537	0.83326	0.00372
72900	0.44865	0.01192	0.81883				0.83075	0.83405	0.01095	0.82978	0.83262	0.00678
73200	0.44964	0.0149	0.82053				0.83543	0.83386	0.01368	0.83421	0.83248	0.00435
73500	0.44985	0.01193	0.82157				0.8335	0.83509	0.01096	0.83253	0.83366	0.00527
73800	0.44897	0.01197	0.82245				0.83442	0.83536	0.01099	0.83344	0.83396	0.00475
74100	0.44973	0.01345	0.8225				0.83595	0.83436	0.01235	0.83485	0.83309	0.00399
74400	0.45005	0.00898	0.82467				0.83365	0.83405	0.00825	0.83292	0.83284	0.00503
74700	0.44948	0.00449	0.82333				0.82782	0.83446	0.00412	0.82745	0.8333	0.00801
75000	0.44993	0.01374	0.82478				0.83852	0.83478	0.01262	0.8374	0.83365	0.00258
75300	0.44954	0.012	0.82586				0.83786	0.83442	0.01102	0.83688	0.83334	0.00285
75600	0.44924	0.0045	0.82498				0.82948	0.83442	0.00413	0.82911	0.83338	0.00708
75900	0.44952	0.00601	0.8271				0.83311	0.83504	0.00552	0.83262	0.83399	0.00515
76200	0.4504	0.00452	0.83075				0.83527	0.835	0.00415	0.8349	0.83397	0.0039
76500	0.44946	0.01202	0.82701				0.83903	0.83574	0.01104	0.83805	0.83469	0.0022
76800	0.44921	0.0045	0.82503				0.82953	0.8354	0.00413	0.82916	0.83441	0.00704
77100	0.45025	0.0075	0.82691				0.83441	0.83493	0.00689	0.8338	0.83401	0.00451
77400	0.44873	0.01358	0.8292				0.84278	0.83518	0.01247	0.84167	0.83426	0.00022
77700	0.4494	0.00601	0.82716				0.83317	0.83541	0.00552	0.83268	0.8345	0.0051
78000	0.45002	0.00752	0.82848				0.836	0.8352	0.00691	0.83539	0.8343	0.00362
78300	0.44975	0.00602	0.82878				0.8348	0.83465	0.00553	0.83431	0.83383	0.0042
78600	0.44902	0.00302	0.82964				0.83266	0.83536	0.00277	0.83241	0.83453	0.00522
78900	0.45004	0.00301	0.82917				0.83218	0.83536	0.00276	0.83193	0.83456	0.00549
79200	0.44962	0.00603	0.82967				0.8357	0.83475	0.00554	0.83521	0.83402	0.0037
79500	0.45008	0.00301	0.82991				0.83292	0.83515	0.00276	0.83267	0.83442	0.00508
79800	0.44984	0.00151	0.83152				0.83303	0.83455	0.00139	0.83291	0.83389	0.00493
80100	0.4498	0.00604	0.83129				0.83733	0.83412	0.00555	0.83684	0.83349	0.0028
80400	0.44974	0.00302	0.83136				0.83438		0.00277	0.83413		0.00427
80700	0.44909	0.00454	0.83158				0.83612		0.00417	0.83575		0.00338
81000	0.44954	0.00604	0.8315				0.83754		0.00555	0.83705		0.00268
81300	0.44983	-0.00151	0.83092				0.82941		-0.00139	0.82953		0.00675
81600	0.44914	0.00151	0.82853				0.83004		0.00139	0.82992		0.00654
81900												
82200												

Table A.4 The combined $(k_R')_{21}$ and $(k_R')_{31}$ spreadsheet for the whole of the third measurement cycle of experiment M7c2

Column 1	The elapsed time/s
Column 2	$10^9 C_P M (\theta - \theta_o) / \int f(\theta) d\tau /WK^{-4}$ Here $\theta_o = 300.3145K$ (the average of the last 11 measurements)
Column 3	$10^9 \int \text{input } d\tau / \int f(\theta) d\tau /WK^{-4}$
Column 4	$10^9 (k_R')_{21} /WK^{-4}$
Column 5	$10^9 (k_R')_{251} /WK^{-4}$, $C_P M /JK^{-1}$, correlation coefficient. The arrows indicate the ranges of the fitting procedures.
Column 6	$10^9 (k_R')_{261} /WK^{-4}$, $C_P M /JK^{-1}$, correlation coefficient. The arrows indicate the ranges of the fitting procedures.
Column 7	$10^9 C_P M (\theta - \theta_o) / \int f(\theta) d\tau /WK^{-4}$ Here $\theta_o = 300.353K$ (the initial value of the cell temperature)
Column 8	$10^9 \int \text{input } d\tau / \int f(\theta) d\tau /WK^{-4}$
Column 9	$10^9 (k_R')_{31} /WK^{-4}$
Column 10	$10^9 (k_R')_{351} /WK^{-4}$, $C_P M /JK^{-1}$, correlation coefficient. The arrows indicate the ranges of the fitting procedures.
Column 11	$10^9 C_P M (\theta - \theta_o) \Delta \int f(\theta) d\tau /WK^{-4}$ (evaluation of $(k_R')_{32}$). Here $\theta_o = 300.3175K$, the average of the 11 measurements preceding the application of the calibration pulse.
Column 12	$10^9 \Delta \int \text{input } d\tau / \Delta \int f(\theta) d\tau /WK^{-4}$ (evaluation of $(k_R')_{32}$)
Column 13	$10^9 (k_R')_{32} /WK^{-4}$

Table A.4

1	2	3	4	5	6	7	8	9	10	11	12	13
300	0.00037	0.82542	0.82579									
600	0.00038	0.82536	0.82574									
900	0.00035	0.8253	0.82566									
1200	0.00038	0.82525	0.82563			0.0036	0.84312	0.83952				
1500	0.00038	0.82519	0.82557			-0.0027	0.84317	0.84587				
1800	0.0004	0.82514	0.82553			0.0012	0.84261	0.84141				
2100	0.00038	0.82508	0.82546			0.0015	0.84263	0.84248				
2400	0.00036	0.82501	0.82537			0.00132	0.84267	0.84135				
2700	0.00038	0.82495	0.82533			0.0001	0.84271	0.84139				
3000	0.00038	0.82488	0.82526			-0.00077	0.84295	0.84372				
3300	0.00036	0.82481	0.82518			0.00008	0.84329	0.84322				
3600	0.00033	0.82476	0.82509			0.00007	0.84362	0.84355				
3900	0.00038	0.8247	0.82508			-0.00054	0.84363	0.84417				
4200	0.00036	0.82465	0.82501			-0.00131	0.84347	0.84478				
4500	0.00039	0.8246	0.82499			-0.0002	0.8433	0.8435				
4800	0.00037	0.82455	0.82491			-0.00065	0.84309	0.84373				
5100	0.00036	0.8245	0.82486			0.00004	0.84282	0.84278				
5400	0.00035	0.82445	0.8248			-0.00036	0.84257	0.84293				
5700	0.00035	0.82439	0.82475			-0.00053	0.84237	0.84289				
6000	0.00036	0.82434	0.82471			-0.00067	0.84222	0.8429				
6300	0.00035	0.8243	0.82464			-0.0003	0.84215	0.84245				
6600	0.00033	0.82424	0.82457			-0.00044	0.84198	0.84243				
6900	0.00036	0.82419	0.82454			-0.00072	0.84179	0.84251				
7200	0.00036	0.82413	0.82449			-0.00097	0.84167	0.84264				
7500	0.00035	0.82408	0.82443			-0.00052	0.84163	0.84215				
7800	0.00034	0.82403	0.82437			-0.0005	0.84159	0.84208				
8100	0.00034	0.82397	0.82431			-0.0006	0.84147	0.84207				
8400	0.00039	0.82392	0.8243			-0.0007	0.84137	0.84206				
8700	0.00034	0.82386	0.8242			-0.00067	0.8413	0.84197				
9000	0.00035	0.8238	0.82415			-0.0002	0.84124	0.84144				
						-0.00062	0.84121	0.84183				
						-0.0006	0.84114	0.84174				

19800	0.00024	0.8217	0.82193				-0.00087	0.83957	0.84044		
20100	0.00024	0.82163	0.82186				-0.00086	0.83956	0.84041		
20400	0.00023	0.82156	0.82178				-0.00089	0.83955	0.84044		
20700	0.00015	0.82149	0.82164				-0.00114	0.83953	0.84067		
21000	0.00017	0.82142	0.82159				-0.00108	0.83951	0.84055		
21300	0.00018	0.82135	0.82153				-0.00102	0.8395	0.84052		
21600	0.00018	0.82127	0.82145				-0.00101	0.83951	0.84052		
21900	0.00022	0.82118	0.8214				-0.00087	0.83955	0.84042		
22200	0.00016	0.8211	0.82126				-0.00106	0.83956	0.84063		
22500	0.00015	0.82103	0.82117				-0.00109	0.83954	0.84063		
22800	0.00023	0.82095	0.82119				-0.00079	0.83954	0.84033		
23100	0.0002	0.82088	0.82108				-0.0009	0.83953	0.84043		
23400	0.00019	0.8208	0.82099				-0.00093	0.83951	0.84044		
23700	0.00014	0.82072	0.82086				-0.00107	0.83952	0.84059		
24000	0.00019	0.82064	0.82083				-0.00091	0.83951	0.84042		
24300	0.00019	0.82057	0.82076				-0.0009	0.83948	0.84037		
24600	0.00014	0.8205	0.82064				-0.00103	0.83944	0.84048		
24900	0.0001	0.82043	0.82053				-0.00113	0.83942	0.84055		
25200	0.00013	0.82035	0.82047				-0.00104	0.83943	0.84047		
25500	0.00014	0.82026	0.82044				-0.001	0.83943	0.84042		
25800	0.00018	0.82019	0.82036				-0.00088	0.83941	0.84029		
26100	0.00014	0.82007	0.82017				-0.00097	0.83948	0.84045		
26400	0.0001	0.81996	0.82006				-0.00107	0.83956	0.84062		
26700	0.00015	0.81988	0.81999				-0.00092	0.83954	0.84045		
27000	0.00013	0.8198	0.81991				-0.00097	0.83952	0.84049		
27300	0.00012	0.81971	0.81983				-0.001	0.83951	0.8405		
27600	0.00012	0.81962	0.81974				-0.00099	0.83951	0.84049		
27900	0.0001	0.81954	0.81964				-0.00101	0.8395	0.84051		
28200	0.00009	0.81945	0.81954				-0.00103	0.83948	0.84051		
28500	0.00009	0.81937	0.81946				-0.00102	0.83946	0.84048		
28800	0.00012	0.81929	0.81941				-0.00094	0.83943	0.84037		
29100	0.00008	0.81921	0.81928				-0.00103	0.83941	0.84044		
29400	0.00008	0.81912	0.81914				-0.00102	0.8394	0.84042		
29700	0.00002	0.81902	0.81904				-0.00113	0.83941	0.84054		
30000	0.00002	0.81892	0.81894				-0.00112	0.83942	0.84053		

40800	0.03996	0.77988	0.81984				0.0466	0.87943	0.83283		0.77535	1.55266	0.77731
41100	0.04094	0.77908	0.82002				0.04686	0.87936	0.8325		0.74495	1.51978	0.77484
41400	0.04173	0.7783	0.82003				0.04688	0.87925	0.83237		0.71379	1.48932	0.77553
41700	0.04263	0.77754	0.82017				0.04701	0.87909	0.83208		0.68662	1.46054	0.77392
42000	0.04354	0.7768	0.82035	0.81802			0.04713	0.8789	0.83177	0.84136	0.66169	1.43369	0.772
42300	0.04444	0.77607	0.82051	469.2			0.04721	0.87869	0.83148	411.2	0.63825	1.40873	0.77049
42600	0.04534	0.77537	0.82071	0.99982			0.04726	0.87842	0.83116	0.9968	0.6163	1.3849	0.7686
42900	0.04614	0.77464	0.82078				0.04719	0.87818	0.83099		0.59443	1.36309	0.76866
43200	0.04692	0.77394	0.82086				0.04709	0.87789	0.8308		0.57377	1.34227	0.76851
43500	0.04776	0.77325	0.821				0.04702	0.87759	0.83057		0.55506	1.32278	0.76772
43800	0.04872	0.77257	0.8213				0.04707	0.87726	0.83019		0.53888	1.30424	0.76537
44100	0.04952	0.7719	0.82142				0.04693	0.87692	0.82999		0.5217	1.28674	0.76504
44400	0.05033	0.77123	0.82156				0.04679	0.87656	0.82978		0.50567	1.2702	0.76453
44700	0.05115	0.77058	0.82173				0.04665	0.87612	0.82954		0.49064	1.25441	0.76377
45000	0.05188	0.76993	0.82181				0.0464	0.8758	0.8294		0.47544	1.23946	0.76402
45300	0.0528	0.76928	0.82208				0.04632	0.87541	0.82909		0.46283	1.22536	0.76252
45600	0.05362	0.76864	0.82257				0.04613	0.875	0.82887		0.4499	1.21186	0.76197
45900	0.05439	0.768	0.8224				0.0459	0.87458	0.82868		0.43714	1.19887	0.76174
46200	0.05525	0.76737	0.82262				0.04571	0.87416	0.82845		0.42563	1.18663	0.761
46500	0.05608	0.76673	0.82281				0.0455	0.87373	0.82824		0.41447	1.17498	0.76052
46800	0.05694	0.76609	0.82303				0.04529	0.8733	0.82801		0.40392	1.1638	0.75988
47100	0.0578	0.76541	0.82321				0.04508	0.87291	0.82783		0.39411	1.15369	0.75958
47400	0.05867	0.76475	0.82343				0.04486	0.87248	0.82762		0.3845	1.14361	0.75911
47700	0.05952	0.76409	0.82361				0.04461	0.87205	0.82745		0.37511	1.13404	0.75893
48000	0.06032	0.76343	0.82375				0.04431	0.87161	0.82731		0.36575	1.12475	0.759
48300	0.06123	0.76277	0.824				0.04408	0.87117	0.82709		0.35737	1.11582	0.75845
48600	0.06221	0.76211	0.82432				0.04389	0.87073	0.82684		0.35883	1.13635	0.77752
48900	0.06297	0.76144	0.82441				0.04354	0.87029	0.82675		0.34102	1.099	0.75799
49200	0.06407	0.76077	0.82484				0.0434	0.86984	0.82644		0.3344	1.09108	0.75668
49500	0.06486	0.7601	0.82496				0.04304	0.86939	0.82634		0.32638	1.08341	0.75702
49800	0.06598	0.75943	0.82541				0.0429	0.86893	0.82603		0.32024	1.07598	0.75574
50100	0.06684	0.75875	0.82559				0.04256	0.86877	0.83921		0.31296	1.06882	0.75586
50400	0.06785	0.75805	0.8259				0.04231	0.86802	0.82572		0.30657	1.06197	0.7554
50700	0.06901	0.75733	0.82634				0.04214	0.86759	0.82544		0.30102	1.05545	0.75444
51000	0.06989	0.75659	0.82648				0.04179	0.86716	0.82537		0.29442	1.04925	0.75482

51300	0.07087	0.75585	0.82672				0.04149	0.86672	0.82523		0.28841	1.04321	0.7548
51600	0.07201	0.7551	0.82711				0.04127	0.86629	0.82502		0.28314	1.03733	0.75419
51900	0.07316	0.75433	0.82749				0.04104	0.86586	0.82482		0.27798	1.03169	0.75372
52200	0.07431	0.75353	0.82784				0.0408	0.86544	0.82464		0.27293	1.02633	0.7534
52500	0.07566	0.7527	0.82836				0.04065	0.86503	0.82438		0.26865	1.02116	0.75251
52800	0.07661	0.75188	0.82849				0.04026	0.86467	0.82435		0.26299	1.01608	0.75309
53100	0.07789	0.75105	0.82894				0.04004	0.86418	0.82414		0.25857	1.01111	0.75255
53400	0.07899	0.75022	0.8292				0.03971	0.86376	0.82404		0.25356	1.00626	0.7527
53700	0.08035	0.74935	0.8297				0.0395	0.86334	0.82384		0.24946	1.0016	0.75215
54000	0.0813	0.74843	0.82973				0.03908	0.86294	0.82386		0.24419	0.99723	0.75304
54300	0.0778	0.75192	0.82972				0.03654	0.86045	0.82391				
54600	0.0745	0.75527	0.82977				0.03419	0.85812	0.82393				
54900	0.07141	0.75849	0.8299				0.03203	0.85594	0.82391				
55200	0.06859	0.76159	0.83018				0.03007	0.8539	0.82383				
55500	0.06563	0.76454	0.83017				0.02813	0.852	0.82387				
55800	0.06301	0.76738	0.83039				0.02641	0.85023	0.82382				
56100	0.06037	0.77011	0.83048				0.02474	0.84857	0.82382				
56400	0.05802	0.77272	0.83074				0.02326	0.84702	0.82375				
56700	0.05567	0.77523	0.83089				0.02183	0.84556	0.82374				
57000	0.05343	0.77764	0.83107				0.02049	0.8442	0.82371				
57300	0.05122	0.77995	0.83117				0.01921	0.84292	0.82371				
57600	0.04914	0.78214	0.83128				0.01803	0.84174	0.82371				
57900	0.04717	0.78424	0.83141				0.01693	0.84063	0.82371				
58200	0.04523	0.78628	0.83151				0.01587	0.83959	0.82371				
58500	0.04349	0.78823	0.83173				0.01493	0.83861	0.82368				
58800	0.04177	0.79011	0.83188				0.01402	0.83769	0.82367				
59100	0.03998	0.7919	0.83188				0.01312	0.83683	0.82371				
59400	0.03848	0.7936	0.83208				0.01235	0.83603	0.82369				
59700	0.03677	0.79524	0.83201				0.01153	0.83528	0.82375				
60000	0.03535	0.79681	0.83216				0.01084	0.83458	0.82374				
60300	0.03385	0.79829	0.83214				0.01014	0.83393	0.82379				
60600	0.03275	0.79969	0.83244				0.00958	0.83332	0.82374				
60900	0.03131	0.80106	0.83237				0.00895	0.83275	0.8238				
61200	0.03	0.80236	0.83236				0.00837	0.83222	0.82385				
61500	0.02877	0.80359	0.83235				0.00784	0.83172	0.82389				

61800	0.02788	0.80475	0.83263				0.00742	0.83126	0.82385		
62100	0.02674	0.80587	0.83262				0.00694	0.83083	0.82389		
62400	0.0257	0.80697	0.83267				0.0065	0.83042	0.82392		
62700	0.02476	0.80805	0.83281				0.00614	0.83003	0.82392		
63000	0.02374	0.80907	0.83282				0.00571	0.82966	0.82396		
63300	0.02296	0.81007	0.83303				0.00538	0.82932	0.82394		
63600	0.02196	0.81103	0.83299				0.00501	0.829	0.82399		
63900	0.02107	0.81193	0.833				0.00467	0.8287	0.82403		
64200	0.02025	0.8128	0.83305				0.00437	0.82842	0.82405		
64500	0.0194	0.81367	0.83306				0.00406	0.82815	0.82409		
64800	0.01861	0.81446	0.83308				0.00379	0.82791	0.82413		
65100	0.01796	0.81521	0.83317				0.00355	0.82769	0.82414		
65400	0.01717	0.81593	0.8331				0.00329	0.82748	0.8242		
65700	0.01668	0.8166	0.83328			0.83403	0.0031	0.82729	0.82419		
66000	0.01583	0.81727	0.8331			510.6	0.00284	0.82711	0.82427		
66300	0.01534	0.81793	0.83327			-0.99908	0.00267	0.82694	0.82427		
66600	0.0149	0.81856	0.83346				0.00251	0.82677	0.82427		
66900	0.01431	0.81918	0.83349				0.00232	0.82662	0.8243		
67200	0.01364	0.81978	0.83343				0.00213	0.82648	0.82435		
67500	0.01319	0.82037	0.83356				0.00199	0.82634	0.82436		
67800	0.01259	0.8209	0.8335				0.00182	0.82623	0.82441		
68100	0.01203	0.8214	0.83344				0.00167	0.82612	0.82445		
68400	0.01158	0.82189	0.83346				0.00153	0.82602	0.82448		
68700	0.01124	0.82238	0.83361				0.00143	0.82592	0.82449		
69000	0.01088	0.82285	0.83373			0.83374	0.00132	0.82583	0.82451		
69300	0.01022	0.82327	0.83349			490.5	0.00117	0.82575	0.82458		
69600	0.01006	0.82365	0.83371			-0.9962	0.00107	0.82569	0.82462		
69900	0.00954	0.82402	0.83356				0.00099	0.82562	0.82464		
70200	0.00941	0.82438	0.83379				0.00099	0.82562	0.82464		
70500	0.00895	0.82474	0.83369				0.00083	0.82551	0.82469		
70800	0.00871	0.8251	0.83381				0.00076	0.82546	0.8247		
71100	0.00854	0.82547	0.83401				0.007	0.82541	0.82471		
71400	0.00784	0.82581	0.83365				0.00058	0.82537	0.82479		
71700	0.00746	0.82607	0.83353				0.0005	0.82534	0.82484		
72000	0.0076	0.82633	0.83393				0.00049	0.82531	0.82482		

72300	0.00746	0.82659	0.83405		0.83314	0.00044	0.82528	0.82484				
72600	0.00703	0.82683	0.83386		440.6	0.00037	0.82526	0.8249				
72900	0.00656	0.82704	0.8336		-0.09821	0.00029	0.82524	0.82496				
73200	0.00648	0.82728	0.83376			0.00026	0.82523	0.82497				
73500	0.00617	0.82755	0.83372			0.0002	0.82521	0.825				
73800	0.00585	0.82779	0.83364			0.00015	0.82519	0.82504				
74100	0.00573	0.82802	0.83375			0.00012	0.82518	0.82506				
74400	0.00536	0.82824	0.8336			0.00007	0.82517	0.82511				
74700	0.00508	0.82843	0.83351			0.00002	0.82517	0.82514				
75000	0.00504	0.82863	0.83367			0	0.82516	0.82516				
75300	0.00513	0.82884	0.83397			-0.00001	0.82516	0.82517				
75600	0.00423	0.829	0.83323			-0.00012	0.82516	0.82528				
75900	0.00444	0.82918	0.83362			-0.00009	0.82516	0.82525				
76200	0.0042	0.82935	0.83355			-0.00012	0.82516	0.82528				
76500	0.00427	0.82937	0.83364			-0.00013	0.82518	0.82531				
76800	0.00417	0.8294	0.83357			-0.00015	0.82519	0.82534				
77100	0.0033	0.82962	0.83292			-0.00021	0.82519	0.8254				
77400	0.00412	0.82986	0.83398			-0.00018	0.82519	0.82537				
77700	0.00269	0.82999	0.83268			-0.00026	0.8252	0.82546				
78000	0.00267	0.83013	0.8328			-0.00027	0.82521	0.82548				
78300	0.00214	0.83032	0.83246			-0.0003	0.82522	0.82552				
78600	0.00178	0.83048	0.83226			-0.00032	0.82523	0.82555				
78900	0.00136	0.8306	0.83196			-0.00034	0.82525	0.82558				
79200	0.00151	0.83073	0.83224			-0.00034	0.82526	0.8256				
79500	0.00094	0.8309	0.83184			-0.00035	0.82527	0.82563				
79800	0.00022	0.83106	0.83128			-0.00037	0.82529	0.82566				
80100	0.00025	0.83111	0.83136			-0.00037	0.82531	0.82568				
80400	-0.0003	0.83106	0.83076			-0.00038	0.82533	0.82571				
80700	-0.00264	0.83099	0.82835			-0.00041	0.82535	0.82575				
81000	-0.00252	0.83083	0.82831			-0.0004	0.82537	0.82576				
81300	-0.00981	0.83047	0.82066			-0.00043	0.82538	0.82582				
81600	-0.0196	0.82973	0.81013			-0.00043	0.8254	0.82584				
81900												

Table A.5 The combined abridged $(k_R')_{21}$ and $(k_R')_{31}$ -spreadsheets prepared according to the instructions in the ICARUS-Systems Handbooks^{(1),(2)} (restriction of the range of the integrations to the region of application of the calibration pulse $t_1 < t < t_2$). The third measurement cycle of experiment M7c2.. The evaluation of $(k_R')_{31}$, $(k_R')_{351}$, $(k_R')_{32}$, $(k_R')_{362}$, $(k_R')_{22}$ and $(k_R')_{262}$. Modification of the procedure for the evaluation of $(k_R')_{22}$ and $(k_R')_{262}$ to take account of the effects of "positive feedback" and evaluation of these coefficients.

Column 1	The elapsed time/s (from the start of the measurement cycle)
Column 2	$10^9 C_{pM} (\theta - \theta_o) / \int f(\theta) d\tau / WK^{-4}$ Here $\theta_o = 300.3175K$, the average of the 11 measurements preceding the application of the calibration pulse
Column 3	$10^9 \int \text{input } d\tau / \int f(\theta) d\tau / WK^{-4}$
Column 4	$10^9 (k_R')_{31} / WK^{-4}$
Column 5	$10^9 (k_R')_{361} / WK^{-4}$, C_{pM}/JK^{-1} , correlation coefficient. The arrows indicate the range of the fitting procedure.
Column 6	$10^9 C_{pM} (\theta - \theta_o) / \Delta \int f(\theta) d\tau / WK^{-4}$ (evaluation of $(k_R')_{32}$ and $(k_R')_{362}$)
Column 7	$10^9 \Delta \int \text{input } d\tau / \Delta \int f(\theta) d\tau / WK^{-4}$ (evaluation of $(k_R')_{32}$ and $(k_R')_{362}$)
Column 8	$10^9 (k_R')_{32} / WK^{-4}$
Column 9	$10^9 (k_R')_{362} / WK^{-4}$, C_{pM}/JK^{-1} , correlation coefficient. The arrows indicate the ranges of the fitting procedures
Column 10	$10^9 C_{pM} (\theta - \theta_o) / \Delta \int f(\theta) d\tau / WK^{-4}$ (evaluation of $(k_R')_{22}$ and $(k_R')_{262}$) Here $\theta_o = 303.0747K$, the average of the last 11 measurements during the application of the calibration pulse.
Column 11	$10^9 \Delta \int \text{input } d\tau / \Delta \int f(\theta) d\tau / WK^{-4}$

- Column 12 $10^9 (k_R')_{22} /WK^{-4}$
- Column 13 $10^9 (k_R')_{262} /WK^{-4}$ C_{PM}/JK^{-1} , correlation coefficient.
The arrows indicate the range of the fitting procedures.
- Column 14 Modification of column 11 to take into account the effects of "positive feedback"
- Column 15 Values of $10^9 (k_R')_{22}$ taking into account the effects of "positive feedback"
- Column 16 $10^9 (k_R')_{262} /WK^{-4}$, C_{PM}/JK^{-1} , correlation coefficient taking into account the effects of "positive feedback".

Table A.5

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
32400																
32700	0.48842	1.27671	0.81029			25.06(903)	24.33(815)	-0.731		0.24271	0.99723	0.75452		1.09389	0.85118	
33000	0.42785	1.25578	0.82793			12.747(14)	13.241(78)	0.494(84)		0.22862	0.98311	0.75449		1.0798	0.85118	
33300	0.40607	1.23659	0.83053			8.5514(0)	9.2018(3)	0.504(3)		0.21588	0.97005	0.75417		1.06681	0.85093	
33600	0.38577	1.21853	0.83375			6.40541	7.1495	0.7441		0.20383	0.95792	0.75409	0.7421	1.05478	0.85095	0.85065
33900	0.364	1.20417	0.84017			5.07334	5.92216	0.84882		0.19286	0.94653	0.75367	477.1	1.04384	0.85098	448.5
34200	0.35352	1.1896	0.83608			4.26961	5.06778	0.79817		0.18338	0.93579	0.75241	0.99897	1.03366	0.85028	0.99998
34500	0.33827	1.17622	0.83795			3.62112	4.44399	0.82287		0.17288	0.9257	0.75281		1.02378	0.8509	
34800	0.32539	1.16323	0.83784			3.15141	3.97478	0.82337		0.16413	0.91609	0.75187		1.0146	0.85047	
35100	0.31181	1.15018	0.83828			2.76989	3.59848	0.82859		0.15565	0.90713	0.75149		1.00624	0.85057	
35400	0.29981	1.13779	0.83798			2.46763	3.29456	0.82693		0.14816	0.89893	0.75076		0.99854	0.85038	
35700	0.29099	1.12608	0.83509			453.6	2.23966	0.84547		0.14104	0.89125	0.75021		0.99113	0.85009	
36000	0.28023	1.11507	0.83484			0.99865	2.03271	0.83835		0.13332	0.88404	0.75073		0.98387	0.85055	
36300	0.27048	1.10475	0.83427			1.66157	2.86515	0.80357		0.12704	0.87724	0.7502		0.97724	0.8502	
36600	0.26116	1.09493	0.83377			1.71403	2.51589	0.80186	0.82987	0.12098	0.87078	0.74881		0.97119	0.8502	
36900	0.25283	1.08563	0.8328			1.58858	2.38598	0.7974	447.2	0.11535	0.86474	0.74939	0.74174	0.96543	0.85008	0.85145
37200	0.24418	1.07686	0.83268			1.47497	2.27325	0.79829	0.89886	0.10976	0.859	0.74924	479.2	0.95999	0.85023	444.5
37500	0.23679	1.06854	0.83175			473.2	1.37989	2.1742	0.79431	0.10497	0.85353	0.74856	0.99885	0.9549	0.84993	0.99995
37800	0.2298	1.06061	0.83081			0.99948	1.28538	2.08589	0.79051	0.09991	0.84836	0.74845		0.94993	0.85002	
38100	0.22279	1.05328	0.8305				1.21766	2.00793	0.79027	0.09507	0.84345	0.74838		0.94514	0.85007	
38400	0.21634	1.04647	0.83014				1.14887	1.93664	0.78976	0.09075	0.83859	0.74784		0.94097	0.85022	
38700	0.20943	1.03973	0.83029				1.0831	1.87497	0.78917	0.08646	0.83378	0.74732		0.93727	0.85081	
39000	0.20398	1.03309	0.82911				1.02949	1.81653	0.78704	0.08312	0.8284	0.74628		0.93352	0.8504	
39300	0.19822	1.02667	0.82844				0.97771	1.76266	0.78495	0.07885	0.82544	0.74658		0.92968	0.85081	
39600	0.19249	1.02061	0.82812				0.92928	1.71383	0.78455	0.07535	0.82176	0.74641		0.92608	0.85073	
39900	0.18757	1.01484	0.82727				0.88753	1.68916	0.78163	0.07231	0.81817	0.74586		0.92279	0.85048	
40200	0.18244	1.00923	0.82679			0.81535	0.8471	1.62754	0.78045	0.06873	0.81472	0.74598	0.74364	0.91952	0.85079	0.85131
40500	0.17786	1.00379	0.82593			480.7	0.81117	1.58878	0.7776	0.06596	0.81154	0.74589	484.2	0.91639	0.85053	445.4
40800	0.17291	0.99855	0.82564			0.99885	0.77535	1.55268	0.77731	0.06286	0.8086	0.74594	0.99924	0.91353	0.85087	0.9997
41100	0.16877	0.99363	0.82485				0.74495	1.51978	0.77484	0.06044	0.80586	0.74542		0.91087	0.85043	
41400	0.16414	0.98893	0.82479				0.71379	1.48932	0.77553	0.05741	0.80311	0.74571		0.90837	0.85096	
41700	0.16015	0.98436	0.82421				0.68662	1.46054	0.77392	0.05563	0.80045	0.74483		0.90611	0.85048	
42000	0.15643	0.97997	0.82355				0.66169	1.43369	0.772	0.05316	0.79802	0.74486		0.90382	0.85066	
42300	0.15281	0.97579	0.82298				0.63825	1.40873	0.77049	0.0506	0.79569	0.7451		0.90134	0.85074	
42600	0.14935	0.97168	0.82232				0.6163	1.3849	0.7686	0.04819	0.79342	0.74523		0.89893	0.85074	
42900	0.14574	0.96787	0.82214				0.59443	1.36309	0.76668	0.04582	0.79145	0.74563		0.89697	0.85115	
43200	0.14223	0.96413	0.82191				0.57377	1.34227	0.76851	0.04417	0.78921	0.74503		0.89502	0.85085	
43500	0.13903	0.96055	0.82153				0.55506	1.32278	0.76772	0.04274	0.78726	0.74452		0.89348	0.85074	
43800	0.13633	0.95708	0.82075				0.53888	1.30424	0.76537	0.04095	0.78535	0.7444		0.89158	0.85063	
44100	0.13325	0.95374	0.82049				0.5217	1.28674	0.76504	0.03817	0.78357	0.7454		0.88944	0.85127	
44400	0.13033	0.95052	0.82018				0.50567	1.2702	0.76453	0.03678	0.78186	0.74508		0.88773	0.85085	
44700	0.12757	0.94738	0.81981				0.49064	1.25441	0.76377	0.03532	0.78022	0.7449		0.88633	0.85101	
45000	0.12465	0.94437	0.81971				0.47544	1.23946	0.76402	0.03378	0.77874	0.74486		0.88519	0.85141	
45300	0.12231	0.94147	0.81916				0.46283	1.22536	0.76252	0.03316	0.77729	0.74413		0.88411	0.85095	
45600	0.1198	0.93868	0.81886				0.4489	1.21186	0.76197	0.03094	0.77587	0.74493		0.8826	0.85166	
45900	0.11727	0.93591	0.81864				0.43714	1.19887	0.76174	0.02865	0.77456	0.74491		0.88128	0.85127	
46200	0.115	0.93329	0.8183				0.42563	1.18663	0.761	0.02684	0.77324	0.74466		0.88029	0.85145	
46500	0.11274	0.93078	0.81802				0.41447	1.17498	0.76052	0.02474	0.77219	0.74479		0.87919	0.85179	
46800	0.11058	0.92829	0.81771				0.40392	1.1638	0.75988	0.02265	0.77103	0.74478		0.87806	0.85181	
							0.40392	1.1638	0.75988	0.02501	0.77003	0.74502		0.87699	0.85198	

47100	0.10854	0.92503	0.81749		0.39411	1.15369	0.75958		0.02365	0.76851	0.74486		0.87534	0.85169
47400	0.10652	0.92378	0.81723		0.3845	1.14361	0.75911		0.02242	0.76735	0.74493		0.87428	0.85186
47700	0.10452	0.92157	0.81705		0.37511	1.13404	0.75893		0.02155	0.76614	0.74459		0.87363	0.85208
48000	0.10248	0.91943	0.81695		0.36575	1.12475	0.759		0.02136	0.76512	0.74377		0.87317	0.85181
48300	0.10067	0.91734	0.81667		0.35737	1.11582	0.75945		0.0201	0.76416	0.74406		0.87222	0.85212
48600	0.09901	0.91532	0.81631		0.35883	1.10635	0.75752		0.01815	0.76324	0.74508		0.8713	0.85315
48900	0.09705	0.91336	0.8163		0.34102	1.099	0.75799		0.01891	0.76238	0.74347		0.87066	0.85175
49200	0.09563	0.91145	0.81582		0.3344	1.09108	0.75668		0.01573	0.76158	0.74585		0.86978	0.85405
49500	0.09378	0.90958	0.81581		0.32838	1.08341	0.75702		0.01676	0.76097	0.7442		0.86896	0.8522
49800	0.09243	0.90778	0.81533		0.32024	1.07598	0.75574		0.01335	0.76048	0.74714		0.86803	0.85468
50100	0.09074	0.90599	0.81525		0.31296	1.06882	0.75588		0.01399	0.7601	0.74811		0.86754	0.85355
50400	0.08926	0.90428	0.81502		0.30657	1.06197	0.7554		0.01267	0.75984	0.74697		0.86667	0.854
50700	0.08801	0.90284	0.81483		0.30102	1.05545	0.75544		0.00898	0.75903	0.75017		0.8653	0.85844
51000	0.08642	0.90109	0.81465		0.29442	1.04925	0.75482		0.01024	0.7582	0.74797		0.86564	0.8554
51300	0.08497	0.89952	0.81455		0.28841	1.04321	0.7548		0.01027	0.75759	0.74733		0.86504	0.85592
51600	0.08373	0.898	0.81428		0.28314	1.03733	0.75419		0.00722	0.75725	0.75003		0.86322	0.8592
51900	0.0825	0.89654	0.81405		0.27798	1.03169	0.75372		0.00402	0.75678	0.75274		0.86019	0.85961
52200	0.08128	0.89514	0.81386		0.27293	1.02633	0.7534		0.00058	0.75579	0.75521		0.85718	0.86733
52500	0.08027	0.89378	0.81351		0.26865	1.02116	0.75251		-0.01015	0.75445	0.7646		0.85731	0.86351
52800	0.07883	0.89244	0.8136		0.26299	1.01608	0.75309		-0.00652	0.75358	0.7601		0.85985	0.87838
53100	0.07775	0.89111	0.81338		0.25857	1.0111	0.75285		-0.01853	0.75349	0.77202		0.86311	0.88601
53400	0.07649	0.88981	0.81332		0.25356	1.00628	0.7527		-0.0229	0.75471	0.77761		0.86599	0.94148
53700	0.07547	0.88855	0.81308		0.24946	1.0016	0.75215		-0.07549	0.75743	0.83293			
54000	0.07409	0.88736	0.81327		0.24419	0.99723	0.75304							

Table A.6 Abridged $(k_R')_{11}$ -spreadsheet for the operation of experiment M7c2 on Day 61.

Column 1	The elapsed time/s
Column 2	The cell temperature/K
Column 3	The cell voltage/V
Column 4	The cell current/A
Column 5	The rate of evaporative cooling/W
Column 6	$f(\theta) / K^4$
Column 7	The "lower bound heat transfer coefficient, $10^9(k_R')_{11} "$ /WK ⁻⁴
Column 8	$10^9(\overline{k_R'})_{11} /WK^{-4}$, σ/WK^{-4} and $\sigma/(\overline{k_R'})_{11} \%$
Column 9	The "lower bound heat transfer coefficient, $10^9(k_R')_{11} "$ /WK ⁻⁴ as given by the evaluation due to the group at the N.H.E. Laboratories
Column 10	$10^9(\overline{k_R'})_{11} /WK^{-4}$, σ/WK^{-4} and $\sigma/(\overline{k_R'})_{11} \%$ of the values of $10^9(k_R')_{11}$ as given by the evaluation due to the group at the N.H.E. Laboratories

Table A.6

[illegible]

9300	344.384	7.3485	1.00079	0.1453	6.47018	0.86536	0.86707	0.86329	0.86509
9600	344.448	7.3253	1.00079	0.1458	6.48125	0.86134	0.00455	0.8594	0.00428
9900	344.483	7.3279	1.00074	0.1461	6.48667	0.86444	0.524	0.86278	0.495
10200	344.517	7.3244	1.00083	0.1464	6.49233	0.86272		0.86102	
10500	344.556	7.3084	1.00081	0.1467	6.49841	0.86262		0.8612	
10800	344.562	7.3097	1.00081	0.1468	6.49949	0.86312	↗	0.86175	↗
11100	344.597	7.3053	1.0008	0.1471	6.50542	0.85884	↗	0.85722	↗
11400	344.627	7.3064	1.00082	0.1473	6.51023	0.86112		0.85974	
11700	344.638	7.2947	1.00079	0.1474	6.51141	0.86189		0.86076	
12000	344.644	7.2896	1.00077	0.1474	6.51291	0.85985		0.85863	
12300	344.664	7.2819	1.00075	0.1476	6.51629	0.85818		0.85695	
12600	344.67	7.2821	1.00077	0.1477	6.51861	0.85698	0.85788	0.85568	0.85665
12900	344.698	7.2746	1.00071	0.1479	6.52185	0.85268	0.00251	0.85114	0.00255
13200	344.727	7.2653	1.00084	0.1482	6.52682	0.85665	0.293	0.85565	0.298
13500	344.703	7.2651	1.00081	0.148	6.52298	0.85862		0.85775	
13800	344.719	7.2681	1.00079	0.1481	6.52602	0.85543		0.85427	
14100	344.723	7.2677	1.00078	0.1481	6.52575	0.85643	↗	0.85536	↗
14400	344.73	7.2631	1.00081	0.1482	6.52679	0.85652	↗	0.85554	↗
14700	344.726	7.2558	1.00081	0.1482	6.52634	0.85568		0.85473	
15000	344.731	7.2502	1.00082	0.1482	6.52736	0.85516		0.85424	
15300	344.723	7.25	1.00082	0.1481	6.52595	0.85694		0.85616	
15600	344.714	7.2451	1.00082	0.1481	6.52437	0.85329		0.85262	
15900	344.73	7.2428	1.00079	0.1482	6.52699	0.85336	0.85483	0.85238	0.85398
16200	344.717	7.2457	1.00084	0.1481	6.52445	0.85707	0.00215	0.85634	0.00225
16500	344.708	7.238	1.00076	0.1481	6.52349	0.85215	0.251	0.8511	0.263
16800	344.728	7.2302	1.00081	0.1483	6.52697	0.85051		0.84847	
17100	344.719	7.2369	1.0008	0.1482	6.52498	0.85491		0.85414	
17400	344.712	7.2315	1.00081	0.1481	6.52363	0.8575	↗	0.85701	↗
17700	344.675	7.2291	1.0008	0.1478	6.51788	0.85839	↗	0.85794	↗
18000	344.664	7.2273	1.00078	0.1477	6.51659	0.85344		0.85256	
18300	344.669	7.224	1.00082	0.1478	6.51669	0.85398		0.8532	
18600	344.649	7.2248	1.00084	0.1476	6.51383	0.85533		0.85462	
18900	344.647	7.2175	1.00082	0.1476	6.51309	0.85498		0.85433	
19200	344.621	7.2277	1.00081	0.1474	6.50955	0.85738	0.85559	0.85677	0.85491
19500	344.616	7.2181	1.00078	0.1473	6.50781	0.85486	0.00192	0.85412	0.00207

19800	344.601	7.2129	1.00079	0.1472	6.50597	0.85594	0.224	0.85535	0.242
20100	344.582	7.216	1.00079	0.147	6.50327	0.85795		0.85746	
20400	344.557	7.2113	1.00081	0.1468	6.49939	0.85721		0.85667	
20700	344.543	7.2077	1.00078	0.1467	6.49689	0.85201		0.85103	
21000	344.561	7.2137	1.0008	0.1468	6.49994	0.85496		0.85419	
21300	344.526	7.2085	1.00079	0.1466	6.4937	0.85801		0.85751	
21600	344.518	7.2027	1.00077	0.1465	6.49218	0.85212		0.85115	
21900	344.528	7.2106	1.00075	0.1466	6.49413	0.85685		0.85624	
22200	344.487	7.2023	1.00083	0.1462	6.48742	0.86098	0.85559	0.85675	0.85451
22500	344.459	7.1998	1.00079	0.146	6.48295	0.8539	0.00239	0.85302	0.00195
22800	344.481	7.2055	1.00069	0.1462	6.48582	0.85417	0.28	0.85328	0.229
23100	344.454	7.2059	1.00055	0.1459	6.48192	0.85571		0.85491	
23400	344.467	7.1974	1.00084	0.1461	6.48405	0.85341		0.85254	
23700	344.448	7.1933	1.00083	0.1459	6.48166	0.85717		0.85664	
24000	344.426	7.1903	1.00083	0.1457	6.47724	0.85419		0.85339	
24300	344.434	7.1955	1.00075	0.1458	6.47855	0.85254		0.85155	
24600	344.431	7.1853	1.00079	0.1458	6.47827	0.85381		0.85307	
24900	344.415	7.1859	1.00075	0.1456	6.47596	0.8549		0.85421	
25200	344.406	7.1834	1.00073	0.1455	6.47416	0.85232		0.8514	
25500	344.411	7.186	1.00065	0.1456	6.4749	0.85613		0.85555	
25800	344.371	7.1859	1.00079	0.1453	6.46908	0.85856	0.85401	0.85811	0.8531
26100	344.363	7.1813	1.00074	0.1452	6.46695	0.85288	0.00202	0.85197	0.00215
26400	344.368	7.1787	1.00071	0.1452	6.4688	0.85082	0.234	0.84978	0.251
26700	344.372	7.1787	1.0006	0.1452	6.46842	0.85379		0.85301	
27000	344.351	7.1773	1.00067	0.1451	6.4652	0.85528		0.85318	
27300	344.356	7.1744	1.00072	0.1451	6.46591	0.85311		0.85229	
27600	344.338	7.1734	1.00076	0.145	6.46277	0.85702		0.85653	
27900	344.312	7.1752	1.00068	0.1447	6.45873	0.8549		0.85415	
28200	344.319	7.1721	1.00066	0.1448	6.46028	0.85128		0.85027	
28500	344.318	7.1645	1.00068	0.1448	6.4595	0.85208		0.85124	
28800	344.309	7.1758	1.00084	0.1447	6.45896	0.85685		0.85625	
29100	344.284	7.1755	1.00078	0.1445	6.45508	0.85451	0.85425	0.85366	0.85348
29400	344.299	7.1679	1.00082	0.1447	6.45629	0.85306	0.00232	0.85221	0.0025
29700	344.275	7.1629	1.00083	0.1445	6.45279	0.85697	0.272	0.85649	0.293
30000	344.254	7.1693	1.00082	0.1443	6.44915	0.85661		0.85596	

30300	344.246	7.1598	1.00081	0.1442	6.44826	0.85305		0.8522	
30600	344.244	7.1589	1.00083	0.1442	6.44824	0.85037	↓	0.84929	↓
30900	344.258	7.1661	1.00082	0.1443	6.45001	0.85413	↑	0.85332	↑
31200	344.231	7.1616	1.00082	0.1441	6.44561	0.85777		0.85729	
31500	344.213	7.1533	1.00081	0.1439	6.44257	0.85295		0.85211	
31800	344.22	7.1489	1.00082	0.144	6.44289	0.85176		0.85088	
32100	344.206	7.1515	1.00083	0.1439	6.44111	0.84997		0.84888	
32400	344.234	7.1622	1.00082	0.1441	6.4461	0.83616	0.85287	0.83375	0.83349
32700	344.347	7.1408	1.00082	0.1453	6.46393	0.85696	0.00576	0.81485	0.01828
33000	344.479	7.1345	1.00081	0.1464	6.48611	0.8537	0.675	0.8118	2.19
33300	344.585	7.1267	1.0008	0.1473	6.50325	0.85643		0.81521	
33600	344.662	7.1168	1.0008	0.1479	6.51616	0.85681		0.81599	
33900	344.736	7.1146	1.0008	0.1485	6.5287	0.85497	↓	0.81425	↓
34200	344.811	7.1122	1.00081	0.1492	6.54068	0.85408	↑	0.81354	↑
34500	344.875	7.0982	1.00082	0.1497	6.55087	0.85249		0.81218	
34800	344.933	7.1005	1.0008	0.1502	6.56029	0.85427		0.81426	
35100	344.973	7.0938	1.00081	0.1506	6.56727	0.85343		0.81357	
35400	345.021	7.0932	1.00079	0.151	6.57464	0.8547		0.8151	
35700	345.04	7.0848	1.00081	0.1512	6.57776	0.85369	0.85292	0.81417	0.81327
36000	345.082	7.0801	1.0008	0.1515	6.58466	0.8543	0.00159	0.81502	0.00151
36300	345.081	7.0767	1.00079	0.1515	6.58501	0.85237	0.186	0.81296	0.185
36600	345.135	7.0705	1.00081	0.152	6.59338	0.84926		0.80982	
36900	345.143	7.0668	1.00082	0.1521	6.59418	0.85075		0.81151	
37200	345.178	7.0659	1.00081	0.1524	6.60096	0.85275	↓	0.81382	↓
37500	345.159	7.0569	1.00081	0.1522	6.59753	0.85175	↑	0.81278	↑
37800	345.195	7.0525	1.0008	0.1525	6.60355	0.85093		0.81206	
38100	345.17	7.0524	1.0008	0.1523	6.59954	0.85465		0.81604	
38400	345.178	7.0512	1.00083	0.1524	6.60117	0.85108		0.81221	
38700	345.181	7.0507	1.00082	0.1524	6.60125	0.85144		0.8126	
39000	345.185	7.043	1.00081	0.1524	6.60181	0.8486	0.85136	0.80962	0.81257
39300	345.202	7.043	1.00079	0.1526	6.6044	0.85129	0.00143	0.8126	0.00151
39600	345.179	7.0462	1.00074	0.1524	6.60072	0.85269	0.167	0.81403	0.186
39900	345.192	7.0403	1.00084	0.1525	6.60275	0.85013		0.81134	
40200	345.182	7.039	1.00083	0.1524	6.60152	0.85146		0.81278	
40500	345.183	7.0423	1.00081	0.1524	6.60148	0.85093	↓	0.81216	↓

40800	345.182	7.0431	1.00081	0.1524	6.6007	0.85115	↑	0.81237	↑
41100	345.183	7.045	1.00083	0.1524	6.60179	0.8537		0.81514	
41400	345.161	7.0433	1.0008	0.1522	6.59817	0.84877		0.81001	
41700	345.205	7.0347	1.00082	0.1526	6.60479	0.8483		0.80946	
42000	345.17	7.0482	1.00081	0.1523	6.59924	0.85542		0.81693	
42300	345.176	7.04	1.00081	0.1524	6.60033	0.8492	0.85135	0.81035	0.81262
42600	345.183	7.0442	1.00082	0.1524	6.60148	0.84975	0.00219	0.81084	0.00231
42900	345.189	7.0407	1.00082	0.1525	6.60277	0.85301	0.258	0.81448	0.285
43200	345.161	7.0456	1.00082	0.1522	6.59806	0.85338		0.81472	
43500	345.176	7.0458	1.00081	0.1524	6.60053	0.85056		0.8117	
43800	345.17	7.0502	1.00073	0.1523	6.59944	0.85165	↓	0.81281	↓
44100	345.182	7.0458	1.00081	0.1524	6.60131	0.85102	↑	0.81221	↑
44400	345.174	7.0504	1.0008	0.1523	6.6001	0.85313		0.81442	
45000	345.194	7.0527	1.00081	0.1524	6.60098	0.84964		0.81068	
45300	345.182	7.0527	1.00078	0.1525	6.60318	0.85145		0.81262	
45600	345.197	7.0447	1.00079	0.1523	6.60142	0.85096		0.81214	
45900	345.207	7.058	1.00083	0.1525	6.60306	0.85026	0.85217	0.81125	0.81332
46200	345.187	7.0628	1.0008	0.1525	6.60573	0.85494	0.00193	0.81593	0.002
46500	345.202	7.0531	1.00082	0.1524	6.60244	0.85467	0.227	0.81595	0.246
46800	345.208	7.0598	1.00073	0.1525	6.60471	0.84976		0.81079	
47100	345.189	7.0678	1.00068	0.1525	6.60528	0.85448		0.81585	
47400	345.217	7.0724	1.00076	0.1524	6.60257	0.85352	↓	0.81466	↓
47700	345.234	7.0707	1.00076	0.1526	6.60707	0.84958	↑	0.81038	↑
48000	345.228	7.055	1.00075	0.1528	6.60977	0.85281		0.81397	
48300	345.242	7.0554	1.0008	0.1527	6.60878	0.85091		0.81208	
48600	345.258	7.0502	1.00081	0.1528	6.6118	0.84811		0.80908	
48900	345.256	7.0425	1.00081	0.153	6.61454	0.84877		0.80992	
49200	345.267	7.0451	1.00082	0.1531	6.6137	0.84826	0.84868	0.80948	0.80978
49500	345.283	7.047	1.00081	0.1532	6.61551	0.84637	0.00184	0.80742	0.00187
49800	345.272	7.0445	1.00074	0.1533	6.61824	0.84878	0.217	0.81006	0.231
50100	345.299	7.0547	1.00074	0.1532	6.61571	0.84744		0.80859	
50400	345.308	7.0516	1.0008	0.1534	6.62078	0.84603		0.80701	
50700	345.309	7.0508	1.00079	0.1535	6.62216	0.84837	↓	0.80962	↓
51000	345.336	7.0575	1.0008	0.1535	6.62232	0.84618	↑	0.80725	↑
				0.1538	6.62605	0.84815		0.80938	

51300	345.324	7.0531	1.00078	0.1537	6.62541	0.84699		0.80816	
51600	345.356	7.0635	1.00082	0.154	6.63006	0.84593		0.80695	
51900	345.362	7.052	1.00083	0.154	6.63136	0.84596		0.80715	
52200	345.377	7.0472	1.00081	0.1542	6.63363	0.85021		0.8055	
52500	345.388	7.056	1.00083	0.1543	6.63523	0.84681	0.84741	0.80811	0.80809
52800	345.391	7.0707	1.0007	0.1543	6.63562	0.8491	0.00191	0.81041	0.002
53100	345.4	7.0565	1.0007	0.1543	6.6368	0.84409	0.226	0.80516	0.248
53400	345.427	7.0657	1.00076	0.1546	6.64176	0.84722		0.80854	
53700	345.415	7.0662	1.00063	0.1545	6.6401	0.85081	↓	0.81242	↓
54000	345.412	7.0603	1.00064	0.1544	6.63929	0.86236	↑	0.82505	↑
54300	345.291	7.0846	1.00071	0.1544	6.61987	0.84558		0.8474	
54600	345.158	7.0774	1.0007	0.1542	6.59726	0.84525		0.84703	
54900	345.056	7.0822	1.00076	0.1543	6.58111	0.84205		0.84345	
55200	344.976	7.0914	1.00073	0.1506	6.56797	0.84282		0.84351	
55500	344.899	7.1001	1.0007	0.1499	6.55522	0.84816	0.84771	0.84905	0.845
55800	344.799	7.1094	1.0007	0.1491	6.53923	0.84719	0.00508	0.84769	0.00656
56100	344.762	7.1233	1.00065	0.1487	6.53234	0.84667	0.599	0.84684	0.776
56400	344.693	7.1181	1.00073	0.1482	6.52134	0.84791		0.84813	
56700	344.652	7.1245	1.00073	0.1478	6.51411	0.84874		0.84886	
57000	344.593	7.1215	1.00072	0.1473	6.50415	0.84803	↓	0.848	↓
57300	344.566	7.1273	1.00072	0.1472	6.50035	0.84713	↑	0.84691	↑
57600	344.527	7.1522	1.00074	0.1468	6.49366	0.85019		0.84981	
57900	344.515	7.1398	1.00077	0.1467	6.492	0.84564		0.84501	
58200	344.501	7.1332	1.0006	0.1465	6.48982	0.84711		0.84667	
58500	344.469	7.139	1.00056	0.1462	6.48407	0.84919		0.84782	
58800	344.459	7.1442	1.00058	0.1462	6.48357	0.84641	0.84816	0.84569	0.84735
59100	344.451	7.1475	1.00059	0.1461	6.48133	0.84573	0.00175	0.84487	0.00164
59400	344.454	7.1603	1.00056	0.1461	6.48151	0.84859	0.206	0.8478	0.193
59700	344.438	7.1725	1.00063	0.146	6.47941	0.8507		0.84992	
60000	344.442	7.1757	1.00052	0.146	6.48017	0.85008		0.8482	
60300	344.442	7.1658	1.00057	0.146	6.48007	0.84895	↓	0.84811	↓
60600	344.435	7.166	1.00061	0.146	6.47954	0.84955	↑	0.84876	↑
60900	344.431	7.1769	1.00063	0.1459	6.47806	0.8466		0.84536	
61200	344.466	7.1781	1.00061	0.1462	6.4842	0.84569		0.84443	
61500	344.464	7.2071	1.00056	0.1462	6.48377	0.85388		0.85295	

61800	344.467	7.1771	1.00057	0.1462	6.48426	0.84815		0.84713	
62100	344.474	7.1762	1.0006	0.1463	6.48571	0.84725	0.84749	0.84619	0.84634
62400	344.482	7.1793	1.00071	0.1464	6.4865	0.84597	0.00242	0.84476	0.00255
62700	344.504	7.1773	1.00062	0.1465	6.49031	0.84438	0.286	0.8431	0.301
63000	344.518	7.1909	1.00061	0.1467	6.49239	0.84605		0.84477	
63300	344.541	7.1979	1.00063	0.1469	6.49626	0.84673		0.84546	
63600	344.554	7.2049	1.00062	0.147	6.49838	0.84808	↓	0.84686	↓
63900	344.572	7.2102	1.00065	0.1471	6.50153	0.84585	↑	0.84438	↑
64200	344.608	7.2215	1.00058	0.1474	6.50784	0.84666		0.84519	
64500	344.626	7.2222	1.00064	0.1476	6.51027	0.8482		0.84689	
64800	344.647	7.2324	1.00073	0.1478	6.51391	0.85095		0.84979	
65100	344.651	7.27	1.0007	0.1476	6.51477	0.85582		0.85455	
65400	344.679	7.2331	1.00068	0.1478	6.51895	0.84772	0.84844	0.84628	0.84709
65700	344.699	7.2255	1.00072	0.148	6.52212	0.84729	0.0033	0.84596	0.0034
66000	344.717	7.226	1.00071	0.1482	6.52548	0.84528	0.389	0.84382	0.402
66300	344.751	7.2306	1.00067	0.1484	6.53167	0.84477		0.84327	
66600	344.772	7.2351	1.00071	0.1486	6.5347	0.84714		0.84582	
66900	344.788	7.2624	1.00068	0.1488	6.53681	0.85317	↓	0.85206	↓
67200	344.79	7.2202	1.00062	0.1488	6.53786	0.84744	↑	0.84641	↑
67500	344.798	7.2237	1.00062	0.1788	6.53814	0.84691		0.84577	
67800	344.809	7.2179	1.00069	0.1489	6.54005	0.84433		0.84306	
68100	344.83	7.2136	1.00068	0.1491	6.54329	0.84401		0.84282	
68400	344.834	7.2136	1.00065	0.1492	6.54446	0.84818		0.84738	
68700	344.817	7.2115	1.0006	0.149	6.54136	0.84698	0.84787	0.84607	0.84694
69000	344.832	7.2132	1.00063	0.1491	6.54423	0.84493	0.00304	0.84384	0.00321
69300	344.832	7.2261	1.00069	0.1491	6.54403	0.84973	0.358	0.84889	0.38
69600	344.823	7.2409	1.0007	0.1491	6.54255	0.85346		0.85273	
69900	344.812	7.194	1.00066	0.149	6.54095	0.84739		0.84675	
70200	344.795	7.225	1.00063	0.1488	6.53816	0.85319	↓	0.8526	↓
70500	344.778	7.2298	1.00062	0.1487	6.53538	0.55315	↑	0.85246	↑
70800	344.771	7.1882	1.0006	0.1486	6.53433	0.84611		0.84534	
71100	344.761	7.2326	1.00059	0.1485	6.53208	0.85309		0.85232	
71400	344.755	7.1932	1.00069	0.1485	6.5315	0.8486		0.84795	
71700	344.733	7.1821	1.00054	0.1483	6.52841	0.84605		0.8453	
72000	344.737	7.2128	1.00054	0.1483	6.52897	0.84946	0.84975	0.8486	0.84902

72300	344.726	7.2202	1.00054	0.1482	6.52655	0.85261	0.00285	0.85189	0.00289
72600	344.715	7.1771	1.00055	0.1481	6.52474	0.84925	0.335	0.8488	0.34
72900	344.678	7.2167	1.00058	0.1478	6.5193	0.85392		0.85328	
73200	344.686	7.1942	1.00038	0.1479	6.5204	0.84645		0.84549	
73500	344.681	7.1901	1.00035	0.1478	6.51979	0.84854		0.8478	
73800	344.666	7.1592	1.00035	0.1477	6.51672	0.84594		0.84536	
74100	344.646	7.1789	1.00065	0.1476	6.51341	0.84805		0.84734	
74400	344.645	7.181	1.00059	0.1476	6.51348	0.84878		0.8481	
74700	344.621	7.1815	1.00054	0.1474	6.50935	0.84938		0.84871	
75000	344.62	7.1957	1.00064	0.1474	6.50918	0.85029		0.84949	
75300	344.608	7.1792	1.0006	0.1473	6.50722	0.8481	0.84863	0.84732	0.84787
75600	344.606	7.1792	1.00053	0.1473	6.50669	0.84788	0.00242	0.84708	0.0024
75900	344.596	7.166	1.00059	0.1472	6.50495	0.84568	0.286	0.84484	0.283
76200	344.598	7.193	1.0007	0.1472	6.50548	0.85113		0.8504	
76500	344.577	7.1615	1.00079	0.1471	6.50307	0.84566		0.84484	
76800	344.588	7.2237	1.00065	0.1472	6.50415	0.85402		0.85313	
77100	344.575	7.1597	1.00065	0.147	6.50161	0.84627		0.84551	
77400	344.571	7.1637	1.00063	0.147	6.50127	0.8468		0.84604	
77700	344.559	7.2123	1.00061	0.1469	6.49961	0.85645		0.85587	
78000	344.538	7.1678	1.00063	0.1466	6.49597	0.84464		0.84514	
78300	344.561	7.1579	1.00064	0.1468	6.50004	0.84599		0.84143	
78600	344.553	7.1853	1.00047	0.1467	6.49863	0.85187	0.8483	0.85123	0.84732
78900	344.532	7.1662	1.00037	0.1465	6.49447	0.84746	0.0035	0.84663	0.00387
79200	344.541	7.169	1.00053	0.1466	6.49636	0.8458	0.412	0.84481	0.457
79500	344.537	7.1705	1.00085	0.1467	6.4957	0.84962		0.84892	
79800	344.518	7.1787	1.00073	0.1465	6.49321	0.85148		0.85081	
80100	344.511	7.1474	1.00064	0.1464	6.49083	0.84494		0.84409	
80400	344.509	7.1462	1.00068	0.1464	6.49112	0.84267		0.84164	
80700	344.52	7.1523	1.00071	0.1466	6.49292	0.84256		0.84147	
81000	344.525	7.154	1.00067	0.1466	6.49374	0.8443		0.84335	
81300	344.522	7.1466	1.00065	0.1464	6.49325	0.84577		0.84503	
81600	344.505	7.1382	1.00055	0.1462	6.49068	0.84511		0.84438	
81900	344.499	7.1419	1.00054	0.1462	6.48959	0.84119	0.8437	0.84006	0.84307
82200	344.522	7.137	1.00057	0.1464	6.49263	0.83925	0.00195	0.83853	0.00194
82500	344.519	7.1427	1.00041	0.1463	6.49235	0.84476	0.231	0.84397	0.23

82800	344.505	7.1504	1.00019	0.1462	6.49068	0.84471		0.84382	
83100	344.513	7.1569	1.00037	0.1463	6.49167	0.84492		0.84395	
83400	344.506	7.1596	1.00046	0.1462	6.49115	0.84549		0.84452	↓
83700	344.514	7.1797	1.00039	0.1463	6.49163	0.84869		0.8485	↑
84000	344.499	7.1619	1.00026	0.1462	6.48939	0.84706		0.84621	↑
84300	344.505	7.1702	1.00034	0.1462	6.49047	0.84954	0.84783	0.84879	0.84702
84600	344.479	7.1514	1.00044	0.146	6.48591	0.84539	0.00266	0.84446	0.00284
84900	344.502	7.1621	1.00041	0.1462	6.48988	0.84265	0.314	0.84139	0.335
85200	344.509	7.1827	1.00041	0.1463	6.49051	0.85151		0.85076	
85500	344.482	7.1735	1.00048	0.146	6.48661	0.85025		0.84945	
85800	344.493	7.1675	1.00048	0.1461	6.48779	0.84753	↓	0.84659	↓
86100	344.48	6.9621	1.00055	0.146	6.48566				
86400	344.49	7.2496	1.00048	0.1461	6.48699				

Table A.7 The operation of experiment M7c2 on Day 68 during the period $0 < t < 21,300\text{s}$ when the cell is driven to dryness. The evaluation given by the group at the N.H.E. Laboratories.

Column 1	The elapsed time/s
Column 2	The temperature measured by the "short thermistor"/K
Column 3	The temperature measured by the "long thermistor"/K
Column 4	The Cell Voltage/V
Column 5	The Cell Current/A
Column 6	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the evaluation by the group at the N.H.E. Laboratories and based on the temperature measurements with the "short thermistor"
Column 7	The rate of excess enthalpy generation, Q_e /W given by the evaluation by the group at the N.H.E. Laboratories and based on the temperature measurements with the "short thermistor"
Column 8	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the evaluation by the group at the N.H.E. Laboratories and based on the temperature measurements with the "long thermistor"
Column 9	The rate of excess enthalpy generation, Q_e /W given by the evaluation by the group at the N.H.E. Laboratories and based on the temperature measurements with the "long thermistor".

Table A.7

1	2	3	4	5	6	7	8	9	
300	370.307	370.362	12.3353	0.9995	0.78377	0.1091	0.77968		
600	370.447	370.498	12.4565	1.0008	0.78779	0.0642	0.78422	0.155	
900	370.551	370.614	12.5636	0.99934	0.78925	0.0479	0.78339	0.1044	
1200	370.688	370.767	12.6606	0.99921	0.78692	0.0743	0.77939	0.1141	
1500	370.837	370.934	12.7268	0.99837	0.78519	0.0941	0.77992	0.1596	
1800	370.936	370.996	12.718	0.99923	0.78006	0.1525	0.77768	0.154	
2100	371.06	371.131	12.8308	0.99987	0.78141	0.1374	0.77487	0.1796	
2400	371.162	371.239	12.9382	0.99955	0.78238	0.1267	0.77746	0.212	
2700	371.293	371.348	12.98	0.99937	0.77767	0.1807	0.77344	0.1829	
3000	371.364	371.436	13.0382	0.99852	0.776	0.2	0.77054	0.2291	
3300	371.498	371.542	13.2664	0.99894	0.78201	0.1317	0.77929	0.2621	
3600	371.602	371.655	13.3062	0.99915	0.77769	0.1814	0.77192	0.1629	
3900	371.711	371.763	13.3877	0.99852	0.77587	0.2027	0.77044	0.2479	
4200	371.78	371.833	13.4814	0.99881	0.77826	0.1754	0.77128	0.2654	
4500	371.872	371.941	13.4839	0.99877	0.76613	0.3155	0.75864	0.256	
4800	371.971	372.017	13.6223	0.99873	0.76835	0.2905	0.76256	0.4024	
5100	372.049	372.12	13.8354	0.99873	0.77579	0.2048	0.7658	0.3577	
5400	372.154	372.21	13.8447	0.99847	0.76409	0.3408	0.75618	0.3207	
5700	372.209	372.281	13.9819	0.99849	0.7705	0.2668	0.75971	0.433	
6000	372.281	372.34	14.02	0.99752	0.76064	0.3817	0.75252	0.3925	
6300	372.363	372.421	14.1844	0.99842	0.765	0.3315	0.75529	0.4765	
6600	372.402	372.463	14.2717	0.99741	0.76782	0.2989	0.75735	0.4449	
6900	372.448	372.507	14.2373	0.99516	0.75616	0.435	0.74706	0.4212	
7200	372.475	372.517	14.3214	0.99917	0.75992	0.3914	0.75045	0.5415	
7500	372.547	372.633	14.4996	0.99784	0.75639	0.433	0.73905	0.5021	
7800	372.64	372.689	14.5917	0.99841	0.74877	0.5228	0.74049	0.6363	
8100	372.678	372.735	14.736	0.99831	0.75423	0.4593	0.74098	0.6201	
8400	372.754	372.814	14.852	0.99863	0.74639	0.5518	0.73293	0.6149	
8700	372.809	372.855	14.9063	0.99783	0.74117	0.6135	0.72944	0.7101	
								0.7516	

9000	372.831	372.895	14.9562	0.99614	0.73977	0.6301	0.72248	0.8338
9300	372.872	372.93	15.1678	0.99634	0.74398	0.5812	0.7294	0.7531
9600	372.943	372.993	15.229	0.9969	0.73054	0.7398	0.71766	0.892
9900	372.982	373.017	15.2966	0.99633	0.72574	0.7968	0.71648	0.9063
10200	373.037	373.069	15.4777	0.99674	0.72328	0.8264	0.7126	0.9528
10500	373.096	373.137	15.668	0.99632	0.72162	0.8469	0.70694	1.0206
10800	373.125	373.163	15.772	0.99609	0.72126	0.8515	0.70762	1.013
11100	373.169	373.209	15.9116	0.9969	0.71639	0.9097	0.70042	1.0984
11400	373.217	373.264	16.1161	0.99656	0.71462	0.9313	0.69572	1.1554
11700	373.265	373.298	16.2655	0.99668	0.708	1.0104	0.69362	1.181
12000	373.306	373.352	16.4239	0.99597	0.7029	1.0714	0.68002	1.3431
12300	373.352	373.403	16.717	0.99439	0.70423	1.0565	0.67864	1.3606
12600	373.401	373.443	16.9188	0.99519	0.69887	1.1209	0.67614	1.3912
12900	373.419	373.467	16.9619	0.99455	0.69384	1.181	0.66744	1.495
13200	373.447	373.478	17.1826	0.99429	0.69508	1.1668	0.67825	1.367
13500	373.488	373.517	17.27	0.99476	0.67749	1.3762	0.65755	1.6136
13800	373.531	373.587	17.5771	0.99437	0.67582	1.3972	0.63664	1.8641
14100	373.568	373.598	17.7841	0.99328	0.66537	1.5221	0.64414	1.7752
14400	373.636	373.682	18.0925	0.99332	0.64023	1.823	0.601	2.2914
14700	373.662	373.691	18.4467	0.99357	0.65036	1.7033	0.62517	2.0041
15000	373.699	373.737	18.6116	0.99374	0.63006	1.9461	0.594	2.3771
15300	373.74	373.755	18.5255	0.9949	0.58375	2.4993	0.56736	2.6953
15600	373.761	373.81	18.7708	0.99327	0.58345	2.5038	0.52564	3.1956
15900	373.781	373.821	18.7899	0.99218	0.56093	2.7731	0.51317	3.3449
16200	373.823	373.875	19.2568	0.99214	0.54855	2.923	0.47861	3.7611
16500	373.859	373.895	19.3422	0.99243	0.51024	3.3822	0.45826	4.0055
16800	373.872	373.916	19.6372	0.99112	0.51522	3.3236	0.44669	4.1453
17100	373.915	373.968	19.9497	0.98994	0.47511	3.8055	0.37976	4.9497
17400	373.924	373.993	20.227	0.99138	0.48689	3.6653	0.35427	5.257
17700	373.924	374.029	20.7173	0.98978	0.52376	3.2246	0.31159	5.7714
18000	373.976	374.091	21.8384	0.9888	0.51714	3.3066	0.23826	6.6566
18300	374.041	374.124	22.8179	0.98688	0.45433	4.0625	0.21433	6.9472
18600	374.08	374.148	23.8283	0.9857	0.43198	4.3332	0.209	7.0142
18900	374.158	374.184	25.8979	0.98207	0.33529	5.5	0.22555	6.8202
19200	374.181	374.213	28.8106	0.9772	0.47236	3.8563	0.31437	5.7566

19500	374.263	374.299	32.9171	0.96991	0.32982	5.5757	0.0357	9.1182
19800	374.366	374.422	38.6512	0.96125	-0.32023	13.4166	-1.55703	28.3434
20100	374.511	374.519	49.9746	0.93923	-7.27982	97.5065	-9.01395	118.4671
20400	374.575	374.673	63.8894	0.90245	129.7446	-1559.12	10.70191	-120.004
20700	374.695	374.788	78.803	0.82992	9.88171	-110.113	7.91021	-86.3652
21000	374.779	374.874	83.3207	0.69007	6.9419	-74.603	6.18571	-65.5346
21300	374.676	375.266	85.5818	0.70753	9.7811	-108.863	5.70622	-60.1171

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Table A.8 The operation of experiment M7c2 on Day 68 during the period $0 < t < 21,300\text{s}$ when the cell is driven to dryness. Evaluation by the ICARUS-Systems methodology but neglecting the effects of reflux D_2O in the cell and with $P^* = 1$ atmosphere.

Column 1	The elapsed time/s
Column 2	The enthalpy input /W
Column 3	The enthalpy change of the calorimeter /W
Column 4	$f(\theta) / \text{K}^4$ as given by measurements with the "short thermistor"
Column 5	The rate of evaporative cooling based on temperature measurements with the "short thermistor"
Column 6	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the ICARUS-Systems evaluation and based on measurements with the "short thermistor"
Column 7	The rate of excess enthalpy generation, Q , /W given by the ICARUS-Systems evaluation and based on measurements with the "short thermistor"
Column 8	$f(\theta) / \text{K}^4$ as given by measurements with the "long thermistor"
Column 9	The rate of evaporative cooling based on temperature measurements with the "long thermistor"
Column 10	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the ICARUS-Systems evaluation and based on temperature measurements with the "long thermistor"
Column 11	The rate of excess enthalpy generation, Q , /W given by the ICARUS-Systems evaluation and based on measurements with the "long thermistor"

Table A.8

1	2	3	4	5	6	7	8	9	10	11
300	10.8029	0.18375	11.20905	1.94411	0.77393	0.85994	11.22023	1.97184	0.77609	0.89718
600	10.93824	0.183	11.23699	2.01878	0.77747	0.82229	11.24736	2.0463	0.77431	0.85863
900	11.01932	0.18075	11.25907	2.07261	0.77857	0.81157	11.2719	2.10845	0.7745	0.85833
1200	11.1248	0.2145	11.28677	2.15177	0.776	0.84256	11.30287	2.20017	0.77061	0.90466
1500	11.18154	0.186	11.31622	2.24289	0.77346	0.87349	11.33601	2.31126	0.76608	0.9587
1800	11.18238	0.16725	11.33735	2.31085	0.76775	0.93984	11.3496	2.35265	0.76324	0.99206
2100	11.30233	0.1695	11.36216	2.40035	0.76856	0.93274	11.37667	2.45361	0.7629	0.99835
2400	11.40607	0.17478	11.38312	2.47677	0.76908	0.92853	11.39887	2.53817	0.76263	1.00333
2700	11.44579	0.1515	11.41003	2.58249	0.76352	0.99414	11.42129	2.62965	0.75864	1.05088
3000	11.49416	0.15373	11.42364	2.64142	0.76149	1.01851	11.43839	2.70623	0.75489	1.09537
3300	11.72696	0.1785	11.45224	2.76564	0.76691	0.95903	11.46126	2.80844	0.76257	1.0095
3600	11.76919	0.15975	11.47296	2.86946	0.76179	1.01949	11.48384	2.92491	0.75624	1.0842
3900	11.84315	0.13352	11.49575	2.98386	0.75905	1.05306	11.50644	3.04247	0.75324	1.1208
4200	11.94017	0.12075	11.5089	3.063	0.76084	1.03363	11.5198	3.12581	0.75467	1.10571
4500	11.94219	0.14323	11.52854	3.17348	0.74819	1.18127	11.54274	3.26151	0.73964	1.28138
4800	12.08993	0.13275	11.54891	3.30112	0.74951	1.16806	11.55839	3.36386	0.74347	1.23882
5100	12.29277	0.13725	11.56487	3.40885	0.75657	1.08799	11.5795	3.51289	0.74637	1.20747
5400	12.29885	0.12	11.58703	3.56395	0.7435	1.24161	11.59857	3.65281	0.73509	1.34029
5700	12.43609	0.09527	11.59899	3.56402	0.75669	1.08988	11.61384	3.77182	0.73783	1.31031
6000	12.46202	0.1155	11.6124	3.76815	0.73873	1.29972	11.62458	3.87265	0.72896	1.41458
6300	12.6374	0.09073	11.62912	3.91843	0.74195	1.26407	11.6411	4.02909	0.73168	1.38492
6600	12.71168	0.06375	11.63749	3.98815	0.74413	1.23964	11.65009	4.10886	0.73296	1.37107
6900	12.64878	0.05476	11.64761	4.06933	0.73188	1.38335	11.65981	4.1909	0.72069	1.51529
7200	12.76378	0.07425	11.65278	4.14073	0.73534	1.34364	11.66146	4.22917	0.72721	1.43946
7500	12.94458	0.12373	11.66746	4.28887	0.73126	1.39295	11.68525	4.48736	0.71316	1.60657
7800	13.04393	0.09825	11.68691	4.50687	0.72207	1.50266	11.69705	4.62902	0.71101	1.63344
8100	13.18668	0.0855	11.69508	4.60059	0.72685	1.44783	11.70689	4.7572	0.71274	1.61449
8400	13.30675	0.09827	11.71051	4.80229	0.71783	1.55536	11.72295	4.97261	0.70254	1.73626
8700	13.35027	0.05776	11.72201	4.954	0.71136	1.63282	11.73155	5.09177	0.69903	1.77871
9000	13.37736	0.04724	11.72575	5.01048	0.70952	1.65487	11.73902	5.22907	0.6901	1.88475

9300	13.59088	0.084	11.73559	5.1369	0.73878	1.3129	11.74762	5.32501	0.69647	1.81125
9600	13.65952	0.08251	11.75021	5.37204	0.69828	1.79035	11.76059	5.54815	0.68269	1.97528
9900	13.71907	0.07049	11.75831	5.5053	0.69256	1.85893	11.76557	5.63349	0.68123	1.99329
10200	13.90522	0.08549	11.76931	5.71171	0.68891	1.90354	11.77596	5.83737	0.67785	2.03487
10500	14.08896	0.066	11.78157	5.94514	0.68563	1.94417	11.79009	6.12047	0.67027	2.12675
10800	14.1893	0.05475	11.7877	6.06672	0.68443	1.95938	11.79559	6.2355	0.66966	2.13487
11100	14.34001	0.069	11.79684	6.26808	0.6784	2.03205	11.80516	6.45768	0.66186	2.22873
11400	14.53891	0.07201	11.80661	6.49473	0.67523	2.07112	11.81639	6.73504	0.65433	2.31976
11700	14.68957	0.06675	11.81629	6.74116	0.66702	2.16987	11.82315	6.9208	0.65144	2.35534
12000	14.83687	0.06524	11.82523	6.96079	0.66052	2.24829	11.8348	7.23059	0.63719	2.52624
12300	15.10478	0.07126	11.83501	7.21912	0.66028	2.25305	11.84563	7.5426	0.63238	2.58557
12600	15.31777	0.05026	11.8449	7.53544	0.65278	2.34379	11.85365	7.82309	0.62803	2.63889
12900	15.35078	0.03449	11.84906	7.65123	0.64689	2.41434	11.85906	7.99215	0.6176	2.76377
13200	15.56621	0.05175	11.85376	7.88956	0.64325	2.4585	11.86022	8.07239	0.62748	2.64683
13500	15.66051	0.063	11.86241	8.15254	0.62761	2.64579	11.86845	8.42626	0.60423	2.92465
13800	15.95974	0.05999	11.8723	8.49419	0.62377	2.69361	11.88397	8.98853	0.58156	3.19788
14100	16.14785	0.07875	11.87898	8.80495	0.61151	2.8407	11.88524	9.08236	0.58785	3.12344
14400	16.45484	0.0705	11.89399	9.45971	0.5822	3.19299	11.90359	9.95922	0.53976	3.70067
14700	16.81091	0.04725	11.89931	9.73827	0.5904	3.09676	11.90537	10.06561	0.56261	3.42925
15000	16.97765	0.0585	11.90714	10.16148	0.56753	3.37114	11.91507	10.63339	0.52755	3.8498
15300	16.91181	0.0465	11.91549	10.68495	0.51868	3.95555	11.91863	10.88498	0.50176	4.15826
15600	17.12775	0.03075	11.91987	10.94911	0.51577	3.99175	11.93011	11.66686	0.45516	4.71821
15900	17.1279	0.04651	11.92344	11.21897	0.49167	4.28026	11.9318	11.8279	0.44029	4.8963
16200	17.59044	0.0585	11.93293	11.85958	0.47535	4.47839	11.9438	12.76064	0.39948	5.38868
16500	17.68034	0.03674	11.94035	12.473	0.43304	4.98645	11.94788	13.14801	0.37627	5.66787
16800	17.94938	0.042	11.94348	12.69194	0.43668	4.94423	11.95268	13.55816	0.36387	5.81833
17100	18.23737	0.039	11.95216	13.52107	0.39134	5.48981	11.96325	14.72758	0.29012	6.70575
17400	18.53881	0.0675	11.95394	13.73194	0.40155	5.3685	11.96838	15.39599	0.26203	7.04483
17700	18.99418	0.0039	11.95405	13.70977	0.44173	4.88821	11.97601	16.40618	0.21577	7.60329
18000	20.08391	0.08775	11.96461	15.0801	0.41088	5.26164	11.98869	18.53509	0.12187	8.73711
18300	21.01156	0.078	11.9776	16.73333	0.35067	5.98851	11.99498	19.8814	0.08772	9.15137
18600	21.98239	0.08775	11.98597	18.05809	0.47409	4.51341	12.00021	20.99736	0.07477	9.3107
18900	23.93393	0.07575	12.00282	21.82352	0.33314	6.21156	12.00827	22.88811	0.08078	9.24481
19200	26.66153	0.07876	12.00785	22.59757	0.33188	6.22928	12.01455	24.63937	0.16175	8.27677
19500	30.44557	0.13875	12.02483	28.46785	0.15293	8.38995	12.03238	32.27347	-0.16345	12.202

19800	35.68564	0.18579	12.04654	42.54893	-0.58517	17.29665	12.0583	58.71612	-1.92535	33.4738
20100	45.50344	0.15675	12.07761	144.1774	-8.18297	109.1045	12.07929	166.8208	-10.0564	131.7494
20400	56.27895	0.138	12.09075	-1636.82			12.11136	-88.2235		
20700	64.1329	0.153	12.11599	-60.3592			12.13557	-34.6757		
21000	56.44338	-0.01425	12.13357	-30.0685			12.15358	-20.7124		
21300	58.05623	-4.02224	12.11261	-60.6579			12.23703	20.97296		

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Table A.9 The operation of experiment M7c2 on day 68 during the period $0 < t < 21,300\text{s}$ when the cell is driven to dryness. Evaluation by the ICARUS-Systems methodology with the inclusion of the effects of reflux of D_2O in the cell. P^* assumed to be equal to the pressure measured at the same time at Sapporo Airport.

Column 1	The elapsed time/s
Column 2	The rate of evaporative cooling /W based on temperature measurements with the "short thermistor"
Column 3	The amount of D_2O evaporated/Mole in each 300s interval corresponding to the rate of evaporative cooling shown in Column 2
Column 4	The amount of D_2O returned to the cell by reflux/Mole corresponding to the rate of evaporative cooling shown in Column 2
Column 5	The amount of D_2O removed from the cell/Mole corresponding to the rate of evaporative cooling shown in Column 2
Column 6	The rate of evaporative cooling /W based on temperature measurements with the "long thermistor"
Column 7	The amount of D_2O evaporated/Mole in each 300s interval corresponding to the rate of evaporative cooling shown in Column 6
Column 8	The amount of D_2O returned to the cell by reflux/Mole corresponding to the rate of evaporative cooling shown in Column 6
Column 9	The amount of D_2O removed from the cell/Mole corresponding to the rate of cooling shown in Column 6
Column 10	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the ICARUS-Systems evaluation and based on measurements with the "long thermistor"
Column 11	The rate of excess enthalpy generation, Q, /W given by the ICARUS-Systems evaluation and based on measurements with the "long thermistor"

Table A.9

[illegible]

9300	5.8477	0.042097	0.00968	0.701569	6.0866	0.043816	0.009982	0.72316	0.74968	1.1862
9600	6.1472	0.044252	0.010161	0.73566	6.3772	0.045908	0.010483	0.758585	0.73602	1.3481
9900	6.3169	0.045474	0.010662	0.770471	6.4850	0.046684	0.011002	0.794267	0.73876	1.3165
10200	6.5827	0.047388	0.011177	0.806682	6.7490	0.048584	0.011529	0.831323	0.73645	1.3449
10500	6.8873	0.04958	0.011715	0.844547	7.1161	0.051228	0.012082	0.870469	0.72816	1.4441
10800	7.0443	0.050711	0.012271	0.882987	7.2652	0.0523	0.012656	0.910113	0.73142	1.4064
11100	7.3082	0.05261	0.01284	0.922757	7.5587	0.054414	0.013244	0.951283	0.72443	1.4901
11400	7.6096	0.05478	0.013429	0.964108	7.9320	0.057101	0.013856	0.994528	0.71593	1.592
11700	7.9402	0.05716	0.014042	1.007226	8.1735	0.058839	0.014494	1.038873	0.71578	1.5946
12000	8.2300	0.059246	0.014682	1.05179	8.5986	0.061899	0.015155	1.085617	0.69949	1.7889
12300	8.5849	0.061801	0.015344	1.098247	9.0243	0.064964	0.015852	1.13473	0.69319	1.8652
12600	9.0127	0.064881	0.016035	1.147093	9.4142	0.067771	0.01658	1.185921	0.6881	1.9268
12900	9.1595	0.065938	0.016754	1.196277	9.6383	0.069384	0.017292	1.238013	0.6813	2.0083
13200	9.4263	0.067858	0.017479	1.246656	9.7332	0.070068	0.018099	1.289982	0.69943	1.7936
13500	9.8483	0.070896	0.018229	1.299323	10.1593	0.073135	0.018872	1.344245	0.67909	2.0362
13800	10.3242	0.074322	0.019015	1.35463	11.0308	0.079408	0.019618	1.404035	0.63903	2.5149
14100	10.7598	0.077458	0.019835	1.412252	11.1458	0.080236	0.020569	1.463702	0.65465	2.3295
14400	11.7469	0.084564	0.020705	1.476111	12.4508	0.089631	0.021477	1.531856	0.58108	3.2089
14700	12.0921	0.087048	0.021651	1.541509	12.5616	0.090428	0.02248	1.599804	0.61526	2.8024
15000	12.6914	0.091363	0.022625	1.610247	13.3905	0.096395	0.023496	1.672704	0.57004	3.343
15300	13.4473	0.096805	0.023651	1.683401	13.6996	0.09862	0.024574	1.74675	0.55203	3.5592
15600	13.8064	0.09939	0.024734	1.758057	14.8843	0.107149	0.025687	1.828212	0.48457	4.3674
15900	14.1481	0.10185	0.025839	1.834069	15.0469	0.10832	0.026889	1.909643	0.48355	4.3801
16200	15.0981	0.108688	0.026977	1.915779	16.4564	0.118467	0.028115	1.999995	0.41708	5.1785
16500	15.9535	0.114846	0.028197	2.002428	16.9372	0.121928	0.029455	2.092468	0.40158	5.3655
16800	16.1975	0.116602	0.02948	2.089551	17.4183	0.125391	0.030829	2.18703	0.39921	5.396
17100	17.3975	0.125241	0.030785	2.184007	19.1407	0.13779	0.032251	2.292569	0.29572	6.6388
17400	17.5276	0.126178	0.032182	2.278003	19.9502	0.143618	0.033822	2.402365	0.27406	6.9008
17700	17.2595	0.124248	0.033566	2.368684	21.2653	0.153084	0.035464	2.519985	0.2214	7.5359
18000	18.9553	0.136456	0.034934	2.470207	24.2559	0.175333	0.03724	2.658079	0.06785	9.3848
18300	21.5679	0.155263	0.03647	2.589	25.5809	0.184152	0.039301	2.802929	0.0677	9.3915
18600	23.1924	0.166957	0.038251	2.717076	26.4748	0.190587	0.041461	2.952055	0.09827	9.0287
18900	27.9715	0.201362	0.040209	2.878859	27.9774	0.201404	0.043696	3.109763	0.16244	8.2642
19200	28.3787	0.204293	0.042611	3.040541	28.5710	0.205677	0.046054	3.269386	0.36699	5.8109
19500	35.2563	0.253803	0.045068	3.249276	35.4106	0.254914	0.04849	3.47581	0.13564	8.6033

19800	49.0363	0.353033	0.048249	3.55403	52.6380	0.378931	0.051663	3.803077	-0.8247	20.237
20100	90.4157	0.650885	0.05291	4.152005	52.7660	0.379852	0.056636	4.126293	0.0371	9.8271
20400	28.4082	0.204505	0.06188	4.29463	48.0871	0.346169	0.061601	4.410861	1.37153	-6.3085
20700	25.4387	0.183128	0.064139	4.413619	21.2551	0.153012	0.065981	4.497892		
21000	16.9942	0.122338	0.066012	4.469946	14.5340	0.104628	0.067383	4.535137		
21300	11.1949	0.08059	0.066739	4.483797	5.5951	0.040246	0.068407	4.506967		

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Table A.10 The operation of experiment M7c2 on day 68 during the period $21,300s < t < 30,300s$ following evaporation to dryness.

Column 1	The elapsed time/s
Column 2	The cell temperature/K
Column 3	The cell voltage/W
Column 4	The cell current/A
Column 5	The enthalpy input /W
Column 6	The rate of change of the enthalpy of the calorimeter /W
Column 7	The rate specific of excess enthalpy generation /W based on the "true heat transfer coefficient, $(k_R')_{12} = 0.85065 \times 10^{-9} \text{ WK}^{-4}$
Column 8	The specific rate of excess enthalpy generation /W based on an assumed "true heat transfer coefficient, $(k_R')_{12} = 0.65 \times 10^{-9} \text{ WK}^{-4}$

Table A.10

[illegible]

Table A.11 The operation of cell M7c2 during the period $2400s < t < 32,400s$ of day 69.

Column 1	The elapsed time/s
Column 2	The cell temperature measured by the "long thermistor" /K
Column 3	The first term on the L.H.S. of equation (A.3). The mean temperature of the water bath was $\theta_{\text{bath}} = 295,204K$
Column 4	The second term on the L.H.S. of equation (A.3)
Column 5	The sum of Columns 3 and 4
Column 6	The rate of change of the enthalpy of the calorimeter /W
Column 7	The rate of enthalpy output from the calorimeter /W. The "true heat transfer coefficient, $(k_R')_{12}$ " has been assumed to be that for a cell filled with gas alone i.e. $0.85065 \times 10^{-9} \text{ WK}^{-4}$
Column 8	The rate of excess enthalpy generation /W

Table A.11

[illegible]

9300	295.316	4.72922	-0.04256	4.68667	0.000989	0.0096	0.0086
9600	295.307	4.81298	-0.04259	4.77039	0.000753	0.0093	0.0085
9900	295.3	4.88335	-0.04261	4.84074	0.000565	0.0077	0.0071
10200	295.295	4.93683	-0.04263	4.8942	0.000471	0.0074	0.0069
10500	295.29	4.99333	-0.04265	4.95069	0.000471	0.0076	0.0071
10800	295.285	5.05322	-0.04266	5.01056	0.000235	0.0075	0.0073
11100	295.285	5.05322	-0.04266	5.01056	0.000235	0.0069	0.0067
11400	295.28	5.11693	-0.04268	5.07425	0.000377	0.007	0.0066
11700	295.277	5.1572	-0.04269	5.11451	0.000235	0.0068	0.0066
12000	295.275	5.18497	-0.0427	5.14228			
12300	295.275	5.18497	-0.0427	5.14228			
12600	295.273	5.21354	-0.0427	5.17084			
12900	295.272	5.22814	-0.04271	5.18543			
13200	295.271	5.24295	-0.04271	5.20024			
13500	295.269	5.27326	-0.04272	5.23054			
13800	295.267	5.3045	-0.04272	5.26178			
14100	295.265	5.33676	-0.04273	5.29403			
14400	295.263	5.37009	-0.04274	5.32736			
14700	295.264	5.35329	-0.04273	5.31056			
15000	295.264	5.35329	-0.04273	5.31056			
15300	295.26	5.42228	-0.04275	5.37953			
15600	295.26	5.42228	-0.04275	5.37953			
15900	295.261	5.40458	-0.04274	5.36183			
16200	295.258	5.45864	-0.04275	5.41584			
16500	295.258	5.45864	-0.04275	5.41584			
16800	295.258	5.45864	-0.04275	5.41584			
17100	295.258	5.45864	-0.04275	5.41589			
17400	295.256	5.49638	-0.04276	5.45362			
17700	295.255	5.51579	-0.04276	5.47303			
18000	295.253	5.5558	-0.04277	5.51303			
18300	295.252	5.57641	-0.04277	5.5762			
18600	295.252	5.57641	-0.04277	5.5762			
18900	295.25	5.61897	-0.04278	5.57619			
19200	295.263	5.37009	-0.04274	5.32736			
19500	295.395	4.19558	-0.04224	4.15329			

19800	295.447	3.95488	-0.04211	3.91277		
20100	295.448	3.95078	-0.04211	3.90867		
20400	295.432	4.01857	-0.04216	3.97641		
20700	295.409	4.12487	-0.04224	3.97633		
21000	295.383	4.26045	-0.04233	4.21812		
21300	295.359	4.40437	-0.04241	4.36196		
21600	295.339	4.54249	-0.04248	4.50001		
21900	295.323	4.66861	-0.04253	4.62608		
22200	295.308	4.80332	-0.04258	4.76073		
22500	295.296	4.9259	-0.04262	4.88328		
22800	295.286	5.04095	-0.04266	4.99829		
23100	295.276	5.17099	-0.04269	5.1283		
23400	295.27	5.25799	-0.04271	5.21528		
23700	295.265	5.33676	-0.04273	5.29403		
24000	295.26	5.42228	-0.04275	5.37953		
24300	295.255	5.51579	-0.04276	5.47303		
24600	295.25	5.61897	-0.04278	5.57619		
24900	295.247	5.68641	-0.04279	5.64361		
25200	295.244	5.75872	-0.0428	5.71592		
25500	295.241	5.83668	-0.04281	5.79387		
25800	295.24	5.86407	-0.04281	5.82126		
26100	295.239	5.89224	-0.04282	5.84943		
26400	295.237	5.95108	-0.04282	5.90826		
26700	295.235	6.0136	-0.04283	5.97077		
27000	295.232	6.11538	-0.04284	6.07253		
27300	295.23	6.18948	-0.04285	6.14663		
27600	295.228	6.26952	-0.04286	6.22666		
27900	295.229	6.2287	-0.04285	6.18585		
28200	295.227	6.31208	-0.04286	6.18584		
28500	295.224	6.45183	-0.04287	6.40897		
28800	295.223	6.50313	-0.04287	6.46025		
29100	295.222	6.55719	-0.04288	6.51432		
29400	295.22	6.67497	-0.04288	6.63209		
29700	295.219	6.73951	-0.04289	6.69662		
30000	295.22	6.67497	-0.04288	6.63209		

30300	295.217	6.88261	-0.04289	6.83971			
30600	295.215	7.04966	-0.0429	7.00676			
30900	295.214	7.14496	-0.0429	7.10206			
31200	295.213	7.25032	-0.04291	7.20742			
31500	295.21	7.65578	-0.04292	7.61287			
31800	295.209	7.8381	-0.04292	7.79518			
32100	295.207	8.34893	-0.04293	8.306			
32400	295.207	8.34893	-0.04293	8.306			
32700	295.42	4.07262	-0.04221	4.03042			

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Table A.12 The final part $76,800s < t < 86,400s$ and the first part $0 < t < 21,300s$ of the operation of experiment M7c2 on Days 25 and 26 respectively

Column 1	The elapsed time/day + 5
Column 2	The temperature measured by the "short thermistor" /K
Column 3	The cell voltage /V
Column 4	The cell current/A
Column 5	The rate of enthalpy input /W
Column 6	The rate of evaporative cooling /W
Column 7	The rate of change of the enthalpy content of the calorimeter /W
Column 8	$f(\theta) /K^4$
Column 9	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴
Column 10	The specific rate of excess enthalpy generation /W cm ⁻³
Column 11	The "lower bound heat transfer coefficient, $10^9(k_R')_{11}$ " /WK ⁻⁴ given by the evaluation of the group at the N.H.E. Laboratories
Column 12	The specific rate of excess enthalpy generation /W cm ⁻³ given by the evaluation of the group at the N.H.E. Laboratories.

Table A.12

[illegible]

85200	334.412	7.6867	0.65385	4.0275	0.0531	0.0111	4.91286	0.80673	1.194	0.80533	-0.307
85500	334.4	7.7094	0.65383	4.0422	0.0531	-0.0103	4.91066	0.81444	0.978	0.81318	-0.51
85800	334.399	7.7171	0.65383	4.0473	0.0531	0.0119	4.90999	0.81106	1.076	0.80966	-0.419
86100	334.415	7.6811	0.65383	4.0237	0.0531	0.0111	4.9127	0.80598	1.214	0.80458	-0.287
86400	334.413	7.6753	0.65383	4.0199	0.0531		4.9125				
Day 26											
300	331.523	8.2294	0.65383	4.3822							
600	330.437	4.2975	0.20097	0.5568	0.0131	-2.1348	4.32896	0.61874			
900	328.826	4.3639	0.20093	0.57	0.012	-2.4548	4.09775	0.73524	1.896	0.75346	3.652
1200	327.337	4.4188	0.20091	0.581	0.011	-2.2753	3.88659	0.73206	1.882	0.74996	0.867
1500	325.952	4.4896	0.2009	0.5952	0.0102	-2.1169	3.69434	0.73136	1.77	0.74882	0.894
1800	324.663	4.5531	0.2009	0.6079	0.0095	-1.9792	3.51663	0.73297	1.708	0.75007	0.872
2100	323.452	4.5967	0.20089	0.6167	0.0089	-1.8446	3.35148	0.73171	1.663	0.74839	0.807
2400	322.333	4.6406	0.20089	0.6255	0.0084	-1.7179	3.20121	0.72942	1.642	0.74561	0.799
2700	321.282	4.7005	0.20088	0.6375	0.0079	-1.6103	3.06152	0.73161	1.542	0.74743	0.81
3000	320.299	4.7275	0.20088	0.6429	0.0075	-1.5144	2.93099	0.73349	1.454	0.74898	0.745
3300	319.369	4.7638	0.20089	0.6502	0.0072	-1.4226	2.8097	0.7352	1.376	0.75032	0.689
3600	318.502	4.7918	0.20088	0.6559	0.0068	-1.3339	2.69709	0.73524	1.329	0.74995	0.641
3900	317.684	4.8229	0.20088	0.6621	0.0065	-1.2524	2.59136	0.73629	1.273	0.75059	0.621
4200	316.92	4.8589	0.20088	0.6693	0.0063	-1.1724	2.49445	0.73583	1.239	0.74969	0.587
4500	316.203	4.8817	0.20088	0.6739	0.006	-1.1044	2.40306	0.7375	1.179	0.75097	0.577
4800	315.525	4.9122	0.20088	0.68	0.0058	-1.1039	2.3179	0.76714	0.747	0.75241	0.54
5100	314.89	4.9339	0.20088	0.6844	0.0056	-0.9809	2.23774	0.74398	0.988	0.75437	0.503
5400	314.286	4.957	0.20088	0.689	0.0054	-0.9254	2.16252	0.74406	1.003	0.75642	0.463
5700	313.721	4.9769	2.0088	0.693	0.0053	-0.8653	2.09327	0.74194	1.004	0.75378	0.424
6000	313.193	5.0034	0.20088	0.6983	0.0051	-0.8178	2.02802	0.74509	0.944	0.75657	0.44
6300	312.688	5.0268	0.20088	0.703	0.005	-0.7703	1.96591	0.74693	0.901	0.75801	0.396
6600	312.22	5.0471	0.20088	0.7071	0.0048	-0.7259	1.9087	0.74827	0.867	0.75897	0.369
6900	311.771	5.0658	0.20088	0.7109	0.0047	-0.6832	1.85467	0.74911	0.84	0.7594	0.348
7200	311.357	5.0743	0.20088	0.7126	0.0046	-0.6436	1.80417	0.74915	0.823	0.75901	0.334
7500	310.958	5.0889	0.20088	0.7155	0.0045	-0.6112	1.7565	0.75274	0.771	0.76227	0.329
7800	310.585	5.111	0.20088	0.72	0.0044	-0.5708	1.711	0.75181	0.767	0.76085	0.29
8100	310.237	5.1165	0.20088	0.7211	0.0043	-0.5407	1.66978	0.75305	0.742	0.76176	0.295
8400	309.902	5.1355	0.20088	0.7249	0.0043	-0.5106	1.62942	0.75504	0.706	0.76397	0.28
8700	309.592	5.1539	0.20088	0.7286	0.0042	-0.4782	1.59237	0.75521	0.699	0.76308	0.254
											0.256

9000	309.298	5.1565	0.20088	0.7291	0.0041	-0.4528	1.55762	0.75601	0.682	0.7637	0.245
9300	309.02	5.1675	0.20088	0.7313	0.004	-0.433	1.52518	0.76076	0.63	0.76805	0.205
9600	308.751	5.1832	0.20088	0.7345	0.004	-0.4014	1.49336	0.75793	0.647	0.76469	0.227
9900	308.513	5.2052	0.20088	0.7389	0.0039	-0.3705	1.46548	0.75433	0.671	0.76056	0.255
10200	308.283	5.2122	0.20088	0.7403	0.0039	-0.3618	1.4389	0.76321	0.589	0.76937	0.183
10500	308.051	5.2182	0.20088	0.7415	0.0038	-0.3467	1.41192	0.76804	0.542	0.774	0.145
10800	307.845	5.2298	0.20088	0.7438	0.0038	-0.319	1.38748	0.76331	0.576	0.76872	0.182
11100	307.653	5.2339	0.20088	0.7446	0.0038	-0.2985	1.36468	0.76161	0.584	0.76664	0.194
11400	307.468	5.2447	0.20088	0.7468	0.0037	-0.281	1.34325	0.76243	0.572	0.7671	0.188
11700	307.298	5.2451	0.20088	0.7469	0.0037	-0.2708	1.32361	0.76606	0.539	0.77058	0.16
12000	307.126	5.2515	0.20088	0.7482	0.0036	-0.2589	1.30345	0.76982	0.506	0.77411	0.134
12300	306.971	5.2636	0.20088	0.7506	0.0036	-0.2351	1.28612	0.76364	0.549	0.76741	0.178
12600	306.829	5.2817	0.20088	0.7542	0.0036	-0.2217	1.26981	0.76631	0.526	0.76922	0.163
12900	306.691	5.2853	0.20088	0.755	0.0036	-0.2185	1.25336	0.77386	0.468	0.77731	0.107
13200	306.553	5.2854	0.20088	0.755	0.0035	-0.2058	1.23786	0.77334	0.463	0.77652	0.111
13500	306.431	5.2905	0.20088	0.756	0.0035	-0.1924	1.2235	0.77229	0.473	0.77515	0.119
13800	306.31	5.295	0.20088	0.7569	0.0035	-0.1805	1.20968	0.77206	0.472	0.77467	0.12
14100	306.203	5.3008	0.20088	0.7581	0.0035	-0.167	1.19708	0.76993	0.484	0.77222	0.135
14400	306.099	5.3114	0.20088	0.7602	0.0034	-0.1615	1.18514	0.77485	0.448	0.77698	0.104
14700	305.999	5.318	0.20088	0.7615	0.0034	-0.156	1.17409	0.77855	0.422	0.78054	0.08
15000	305.902	5.3148	0.20088	0.7609	0.0034	-0.148	1.16277	0.77876	0.418	0.78054	0.079
15300	305.812	5.317	0.20088	0.7613	0.0034	-0.1346	1.15257	0.77439	0.446	0.77587	0.107
15600	305.732	5.3123	0.20088	0.7604	0.0034	-0.1243	1.14322	0.77091	0.467	0.77212	0.129
15900	305.655	5.3231	0.20088	0.7626	0.0034	-0.1195	1.13473	0.77442	0.442	0.77547	0.108
16200	305.581	5.3234	0.20088	0.7626	0.0033	-0.1195	1.12607	0.78043	0.402	0.78153	0.071
16500	305.504	5.3312	0.20088	0.7642	0.0033	-0.1077	1.1176	0.77715	0.422	0.77788	0.092
16800	305.445	5.327	0.20088	0.7633	0.0033	-0.1021	1.11066	0.77626	0.426	0.77684	0.098
17100	305.375	5.3423	0.20088	0.7664	0.0033	-0.0998	1.10248	0.78266	0.384	0.78321	0.06
17400	305.319	5.3297	0.20088	0.7639	0.0033	-0.0903	1.09652	0.77596	0.426	0.77624	0.1
17700	305.261	5.3285	0.20088	0.7637	0.0033	-0.0823	1.08981	0.77326	0.386	0.77327	0.116
18000	305.215	5.3391	0.20088	0.7658	0.0033	-0.0776	1.08427	0.7748	0.431	0.77468	0.108
18300	305.163	5.3537	0.20088	0.7687	0.0033	-0.0752	1.07826	0.77965	0.401	0.77946	0.08
18600	305.12	5.3425	0.20088	0.7665	0.0033	-0.0713	1.07337	0.77742	0.413	0.77711	0.093
18900	305.073	5.3408	0.20088	0.7661	0.0032	-0.0689	1.06793	0.77886	0.403	0.77847	0.085
19200	305.033	5.3493	0.20088	0.7678	0.0032	-0.0657	1.06308	0.78103	0.389	0.78056	0.073

19500	304.99	5.3485	0.20088	0.7677	0.0032	-0.0673	1.05861	0.78568	0.36	0.78525	0.046
19800	304.948	5.3525	0.20088	0.7685	0.0032	-0.0578	1.05477	0.78031	0.392	0.77959	0.078
20100	304.917	5.3595	0.20088	0.7699	0.0032	-0.0515	1.05084	0.77854	0.403	0.77766	0.088
20400	304.883	5.3617	0.20088	0.7703	0.0032	-0.0523	1.04709	0.78252	0.378	0.78162	0.066
20700	304.851	5.3547	0.20088	0.7689	0.0032	-0.057	1.04315	0.78868	0.34	0.78793	0.031
21000	304.811	5.3554	0.20088	0.7691	0.0032	-0.0491	1.03852	0.78471	0.364	0.78372	0.054
21300	304.789	5.3659	0.20088	0.7712	0.0032	-0.0348	1.03644	0.77458	0.425	0.77311	0.112