



Research Article

Excerpts from Martin Fleischmann Letters

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Abstract

There were many letters exchanged between Martin Fleischmann and this author that reveal Fleischmann's scientific thoughts about various cold fusion topics. These topics included possible critical factors for excess enthalpy effects in the palladium/deuterium system, errors in the CalTech, MIT and Harwell calorimetry, helium-4 production, critics such as Morrison, Taubes and Jones, and the advantages of using integrated forms of the calorimetric equations. Furthermore, Fleischmann mathematically shows that there is never a true steady state in the cell temperature for an open isoperibolic calorimetric system because both the cell heat transfer coefficient and the heat capacity of the cell change with time. Therefore, an important power term involving the change of the cell temperature with time has often been neglected by various groups using isoperibolic calorimetry. This power term is especially important when there are large changes in the cell temperature with time such as when the cell is first turned on.

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Keywords: Calorimetry, Cell temperature, Critics, Excess enthalpy, Helium

1. Introduction

Martin Fleischmann was an outstanding scientist with a detailed grasp of chemistry, physics and mathematics that is seldom found in one individual. This is shown in a recent book about areas of science inspired by Martin Fleischmann [1]. Cold fusion is just one of the 19 chapters in this book on various topics, where Fleischmann made major contribution [1]. Eventually, Fleischmann's genius in cold fusion will be recognized by mainstream science despite the brutal criticism that he endured along with Stanley Pons. Fleischmann and Pons will then be in good company with Galileo, Joule, Semmelweis and other great scientists whose accomplishments were very slow in becoming accepted.

Fleischmann knew that James Prescott Joule had similar problems in gaining acceptance for his work some 150 years earlier. Joule studied the nature of heat and its relationship to work which led to the development of the first law of thermodynamics. At one point, Joule had to present his ideas in a church because they were rejected by his fellow scientists. In 1999, when there was much opposition to a cold fusion session that I had organized for an American Chemical Society (ACS) meeting in Ontario, California, Fleischmann proposed that we follow Joule's example and hold our session in a church. Fortunately, cooler heads eventually prevailed and our cold fusion session was permitted at this ACS meeting. However, we were moved to a room at a remote corner of the building.

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Only a small portion of these Fleischmann letters can be presented here, but a book is in preparation by the author that will present fully many of these letters. It is hoped that these letters will show Fleischmann's character and thoughts on various topics during the early history of cold fusion.

2. Possible Critical Factors for Cold Fusion

Prior to their 1989 announcement, the Fleischmann and Pons cold fusion work was funded by the US Office of Naval Research (ONR), managed first by Jerry Smith and later by Bob Nowak. I was also funded at China Lake by ONR for several years that included research in 1991 to test palladium materials made by the Naval Research Laboratory (NRL). None of these electrodes produced any evidence for the anomalous excess heat effect that I had previously observed using palladium rods purchased from Johnson-Matthey. In frustration, I sent Bob Nowak a list of questions to give to Fleischmann about possible critical factors for producing excess heat in the palladium/deuterium system. Fleischmann's answers to my questions were included with other topics in a confidential letter dated 21 January 1992. The following shows my 10 questions followed by Fleischmann's answers:

- (1) *Heat treatment of palladium prior to the experiment?* "We do not heat treat the palladium prior to the experiment".
- (2) *Anodic treatment of palladium prior to deuterium loading?* "We do not polarize the electrodes anodically prior to making calorimetric measurements. However, we have now carried out extensive investigations on the factors which control the deuterium loading and this includes anodic treatment. I wished I could say that we understand the complexities of this topic but in truth we do not".
- (3) *Current density to use for deuterium loading?* "We always load at low to intermediate current densities, which is less than 100 mA cm^{-2} before we raise the current density. We believe this to be a critically important point and to the best of our knowledge, there are only three groups of people in the world who understand the reason for doing so which are Stan and I, EPRI/SRI and the National Fusion Centre in Nagoya.
The reason why we have talked about this matter the least is that our protocol is based on a surmise and we do not want to polarize people's thoughts. Equally, it affects the whole of our research strategy".
- (4) *Effect, if any, of atmospheric CO_2 ?* "I do not know what the effects of atmospheric CO_2 might be. At the very least, it will complicate the calorimetry if CO_2 gets reduced at the cathode".
- (5) *Effects of D_2O impurities such as Cu, H_2O ?* "The impurities in the D_2O are probably of key importance and this goes for metals, as well as the borates and silicates. I wished I could tell you that we had this sewn up but we have not. However, we do believe that the blocking of the surface either by Under Potential Deposition (UPD) layers or by insoluble precipitates is very important. As you will see, I am sending a copy of this letter to Bob so I would just expand my comments here by telling you that it is Stan's and my view that as far as the bulk of the palladium is concerned, it is advantageous to produce a set of microelectrodes by suitable blocking of the surface".
- (6) *Effects of impurities in lithium metal such as Hg, Sn, Zn, and Pb?* "The third impurity on your list gets into the D_2O anyway and we believe that this is one of the species which is implicated!"
- (7) *Effects of sudden voltage or current changes?* "Sudden changes in the electrochemistry are very important. You will want to know that our strategy in Utah was oriented at developing a reproducible set of electrodes and cells on which we were then going to investigate systematically the effects of perturbations in temperature and perturbations in the electrochemistry. Unfortunately, our work there was terminated because the people at National Cold Fusion Institute (NCFI) did not buy any more D_2O . This is one topic which we have to start anew".
- (8) *Effect of bubble patterns at the cathode?* "There is clearly a wide variation in the formation of bubbles at the cathode. If bubble evolution is irreproducible then this leads to noise in the electrode potential: the noise

levels for different cells are widely variable. You will be interested to know that this affects our data evaluation because the errors in fitting the black box model to the data can become totally dominated by the fluctuations in cell potential, rather than the time dependent change of temperature. We believe that Kalman filtering is therefore a better technique to use than the non-linear regression technique which we have so far used extensively”.

- (9) *Effect of impurities/additives in Pd such as Ce, Li, or Ag?* “I am sure you will know that our favored strategy now is to use alloys (which you have listed! – no prizes for guessing)”.
- (10) *Effect of Li-6 versus Li-7?* “We do not know this as yet.”

After responding to my 10 questions, Fleischmann made some additional comments: “I would like to make some more general comments with regard to electrode materials. You will know that in our original measurements with palladium alone, we got good reproducibility with 1, 2 and 4 mm diameter rods but never had any excess heat with our original 8 mm rods. We believe that this showed Johnson Matthey that cold working was very important and in fact, we also believe that large diameter electrodes “do not work” because they crack. We have subsequently managed to get excess heat with specially prepared 8 mm rods (I presume that Johnson Matthey changed their procedures so that the rods were prepared in an analogous way to those having smaller diameters but our measurements with these rods proved to be irreproducible).

It is also true that a subsequent batch of 1, 2 and 4 mm diameter rods gave us irreproducibility and low levels of excess heat but we managed to recover more or less our original position with yet further batches of electrode materials.

Our promising results with alloys have been based on single batches of material and in view of our experience with the palladium electrodes, we now feel that we have to carry out a major investigation with repeated batches of alloy electrodes. Bob (Nowak) will tell you that Stan and I are in favour of making these materials widely available if and when we find that they give us reproducible results (but the decision is not up to us).

There is another important point which I must raise with you and that is most of our results are based on the first charging of the electrode. You may recall that at the Como conference, I said that people should give this information in future research. As of now, we do not know whether the first charge or any subsequent charge of the electrode will be optional. This information is obviously critically important but it will take several years to decide this point. I only hope that someone somewhere will provide the resources so that these long-term experiments can be initiated and maintained”.

Note: Regarding Fleischmann’s last comment above, several electrodes that gave excess heat in my experiments were cleaned and polished and then gave similar large excess heat effects in new experiments conducted both at China Lake and at the New Hydrogen Energy laboratory (NHE) in Sapporo, Japan.

3. Helium-4 Production in the Pd/D System

Fleischmann and Pons were actually the first to observe that helium-4 was produced in the Pd/D system. This was mentioned to me by Fleischmann in several conversations as well as in a letter dated 21 September 1993. Although this letter was mainly about other issues, including my problems with Steve Jones, Fleischmann states: “Next your Steve Jones saga. Of course he is terrified that people will come to accept heat and ^4He (incidentally, we had our first indication of ^4He in December 1988!).” In conversations with me, Fleischmann stated that because of the intense criticism following their March 23, 1989 announcement, they did not want to open up a second front about helium-4 production.

At a SPAWAR (Navy) meeting in San Diego in 2000, Fleischmann mentioned an experiment in France that produced excess heat for several months. According to Fleischmann, the excess heat production eventually ended because the palladium lattice became choked with helium-4. Fleischmann always considered that the excess heat in the Pd–D system was “a bulk effect mediated by surface reactions.” My communications with both Fleischmann and Pons sug-

gests that considerable work on helium-4 production was performed at their laboratory in France (IMRA Europe). It is hoped that the results of this research will someday be released.

My China Lake correlation of heat and helium was performed in the fall of 1990 with results first reported in 1991 [2,3]. I was not aware at this time of Fleischmann and Pons results for helium-4. In fact, I was expecting possible helium-3 based on Schwinger's suggestion of the $D+H \rightarrow He-3$ reaction that produces no neutrons or gammas. However, only helium-4 was detected. These experiments were conducted entirely at China Lake except for the helium-4 measurements on the gas samples sent to the University of Texas. I allowed the initial paper [3] to be released through the University of Texas because I was concerned about enemies of cold fusion research at China Lake who may have blocked this publication. Further research at China Lake confirmed our earlier heat/helium results [4–6].

Research by others followed that also showed helium-4 production in the Pd/D system. This includes results of McKubre, Gozzi, DeNinno, Arata and others [7]. In my opinion, any correct theory for cold fusion in the Pd/D system would have to explain the production of helium-4, and many theories do this. However, new theories are sometimes proposed that apparently consider the production of helium-4 to be some mistake [8]. After all these years, I do not find any errors in my heat and helium measurements. It is very unlikely statistically that my heat-helium results could have been produced by random errors [5,6].

4. Comments About Cold Fusion Critics

The most vocal critics following the 1989 announcement included Douglas Morrison, Gary Taubes and Steven Jones. Especially harsh personal statements were also made very early after the announcement by Nate Lewis and Steve Koonin of CalTech [9]. However, Fleischmann never expressed anger in any of his comments about these critics. Only a few comments from my Fleischmann letters will be given here.

4.1. Douglas R.O. Morrison

Douglas Morrison of CERN attended most of the early ICCF conferences and wrote biased reports about cold fusion that were sent to many scientists. The Fleischmann's letters of April and May of 1992 discussed possible ways to respond to Morrison's "Cold Fusion update No. 6" dated 5 April 1992. Fleischmann wanted to respond to Morrison in an accepted scientific manner and stated in one letter: "And come on Douglas: Write some papers for peer-reviewed journals". Morrison eventually submitted a paper to Physics Letters A, but this was rejected by the editors. Fleischmann sent me a copy of this submitted Morrison paper titled "Comments on Claims of Excess Enthalpy by Fleischmann and Pons Using Simple Cells Made to Boil". Fleischmann also sent me their reply to this Morrison paper that was included with his 21 September 1993 letter. Fleischmann stated in this letter: "With hindsight the outcome was predictable: the editors refused to accept Douglas Morrison's critique which they regarded as being too polemical and asked him to submit a shortened non-polemical comment. Naturally, this has once again deprived us of a "right to reply". (Note: These two papers have now been published: see *Infinite Energy* 21(124) (2015) 17–27.)

4.2. Gary Taubes

In an article about Fleischmann's cold fusion seminar in 1991 at CalTech, Gary Taubes made some false statements about my China Lake helium-4 results (Science, News and Comments, 13 December, p. 1582, 1991). The editors limited my reply to a letter of only two paragraphs (Science, Letters, 13 March, p. 1335, 1992). Fleischmann states in his letter of 23 March 1992: "I find it really remarkable that people like Gary Taubes can have access to such Journals with uncorroborated statements (others might say lies) while reasoned arguments do not prevail". I had previously sent Fleischmann my full reply that covered several important topics, but only a small portion was published.

4.3. Steven Jones

Fleischmann obviously did not trust Steven Jones of Brigham Young University (BYU), and thought that Jones acted as a reviewer on a 1988 Fleischmann–Pons proposal and then used the information in the proposal to advance his own cold fusion experiments. Fleischmann made the following comments in his 21 September 1993 letter: “He (Jones) has a total blockage about the excess enthalpy measurements because of his disbelief of our results in 1988/Spring 1989 that precipitated the unfortunate chain of events leading to the Press Conference”. Fleischmann also stated the following about the referee’s comment: “Did we ever tell you what happened when we had the referees’ comments on our application? I said to Stan: ‘Hey Stan, this is Steven Jones and if we answer question X, then we tell him why there may be fusion in the earth and more likely Jupiter; if we answer question Y, we tell him how to set it up in the laboratory: Prophetic words.’ I also had problems with Steven Jones becoming a major critic of my work after inviting me to give a seminar at BYU that lasted more than three hours due to his many probing questions.

5. CalTech, Harwell and MIT Calorimetry

The failure of CalTech, Harwell and MIT to report any evidence for excess heat in their 1989 experiments on the Pd/D system was a major setback for the acceptance of cold fusion. In a 10 April 1992 letter, Fleischmann stated that the cold fusion publications from these institutions “were very influential in creating a negative climate of opinion – not because of their excellence but because of the reputations of CalTech, Harwell and MIT.” In a 1 March 1995 letter, Fleischmann states “that the trio of results at MIT, CalTech and Harwell still pose a tremendous block to further work.” In several letters in 1992, Fleischmann was trying to have a committee of scientists set up to carefully examine the calorimetry of various groups including that of Fleischmann and Pons along with CalTech, Harwell, and MIT. If this examination were carried out correctly, Fleischmann knew that his calorimetry would prove to be superior. Unfortunately, such a committee was never established. However, major errors for the CalTech, Harwell, and MIT calorimetry have been reported [10–12]. Furthermore, these three groups never reported obtaining the high D/Pd ratio required for producing excess heat or any other cold fusion effects [1].

Fleischmann’s detailed letter of 2 April 1992 states: “As you will see we are in favour of reviewing the published material and we do have a set of questions which we believe should be considered”. This letter lists 12 questions for the CalTech calorimetric papers and 29 questions for the Harwell calorimetry. These questions refer to possible errors based on the published papers.

One example of a calorimetric error will be presented here. An important term missed by CalTech, Harwell and MIT [10,11] is the power due to the rate of change of the cell temperature with time: $P_{\text{calor}} = C_p M dT/dt$, where $C_p M$ gives the heat capacity of the calorimeter (JK^{-1}) and dT/dt gives the change in the cell temperature with time (Ks^{-1}). This term is especially important when the electrolysis is first initiated, when there is an applied internal heater, when there is a change in the applied cell current, when D_2O is added and when the cell is turned off. These important periods can be accurately modelled for isoperibolic calorimetry using $P_{\text{calor}} = C_p M dT/dt$ and should never be neglected. Several of Fleischmann’s questions of the William’s paper [13] relate to the strange endothermic behavior shown in William’s Figs. 2(b–e) on p. 379. For example, Fleischmann’s Question 18 for Fig. 2(e) in the William’s paper states: “Why was the excess enthalpy for the dissolution of D in Pd not seen and what could be the cause of the large negative enthalpy over the first two hours?” The answer is William’s neglect of the P_{calor} term which would be large and positive when the cell is turned on and the contents heat up.

This same error in neglecting P_{calor} has also frequently been made by noted cold fusion scientists. For example, Storms erroneously states that the temperature of an isoperibolic cell needs to be constant before the values have any meaning [14]. Actually, there is never a steady state where $dT/dt = 0$ for an open isoperibolic calorimeter, thus the P_{calor} term is always required. The positive and negative false excess power results shown in several of Storm’s figures are due to the neglect of this P_{calor} term [14].

6. Temperature Versus Time for Isoperibolic Calorimetry

Fleischmann was foremost a consummate mathematician in developing models of electrochemical and other systems. Therefore, we must briefly enter this world. A long Fax was received from Stan Pons on 8 July 1992 that contained 27 pages of Fleischmann's modeling equations. In regard to Fleischmann's notes, Stan stated: "It should be possible to predict the temperature–time data for whole sequences of measurements (over periods of months) and for various modes of operation (including the Harwell type of experiments). This type of modelling should also really facilitate data evaluation and improve the accuracy."

Fleischmann's model of his open isoperibolic calorimetry where the electrolysis gases (D_2 , O_2) escape along with D_2O vapor is given by the differential equation

$$d\Delta\theta/dt + (\gamma - yt)\Delta\theta/(1 - \beta t) = \alpha/(1 - \beta t). \quad (1)$$

Note that $\Delta\theta = T - T_b$, thus $d\Delta\theta/dt = dT/dt$ if the bath temperature is constant. All other terms are defined in the Appendix. If the heat transfer coefficient and $C_p M$ are assumed to be constant, then both $y = 0$ and $\beta = 0$. Eq. (1) then becomes

$$d\Delta\theta/dt + \gamma\Delta\theta = \alpha. \quad (2)$$

Equation (2) can be readily integrated to give

$$\Delta\theta = (\alpha/\gamma)[1 - e^{-\gamma t}]. \quad (3)$$

If only the heat transfer coefficient is assumed to be constant, then only $y = 0$ and Eq. (1) becomes

$$d\Delta\theta/dt + \gamma\Delta\theta/(1 - \beta t) = \alpha/(1 - \beta t). \quad (4)$$

This equation can also be integrated using known integrals to yield

$$\Delta\theta = (\alpha/\gamma)[1 - (1 - \beta t)^{\gamma/\beta}]. \quad (5)$$

However, neither the cell heat transfer coefficient nor the cell heat capacity remains constant in the open isoperibolic calorimeter used by Fleischman and Pons, thus Eq. (1) must be integrated in order to obtain the correct cell temperature–time behavior. There are no known integrals for Eq. (1), and Fleischmann's note contains many pages of mathematics to obtain the integrated equation for the correct temperature–time behavior. Fleischmann's final integrated solution to Eq. (1) is given by

$$\Delta\theta = \alpha[(\gamma - yt)^{-1} - \gamma^{-1}(1 - \beta t)^{\varphi} e^{-yt/\beta}], \quad (6)$$

where $\varphi = \beta^2(y - \gamma\beta)^{-1}$.

Equations (3), (5) and (6) all show that the cell temperature increases nearly linearly with time, $\Delta\theta = \alpha t$, when t is less than about 1000 s. The correct equation, Eq. (6), shows that there is never a true steady state for the cell temperature even over long time periods.

Figure 1 shows the cell temperature versus time for the first three hours for Eqs. (3), (5) and (6). The initial increase in the cell temperature is nearly linear with time with the slope = α . The differences between these three equations are small over the first three hours and undetectable in Fig. 1.

The results are quite different for the first eleven days as shown in Fig. 2. The black line shows the correct behaviour corresponding to Eq. (6). There is never a steady state where the cell temperature remains constant. In most experiments, the cell would be refilled every two or three days (3 days = 259,200 s).

Although the results for Eqs. (3) and (5) shown in Fig. 2 are very similar, Eq. (5) never yields a true steady-state temperature. The difference between the cell and bath temperature, $\Delta\theta$, can eventually become zero or even negative

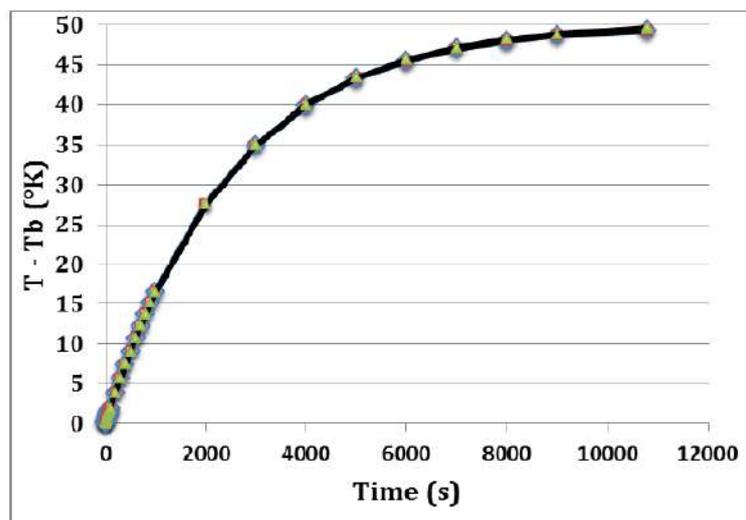


Figure 1. Cell temperature versus time for the first three hours of cell operation based on Eqs. (3), (5) and (6). The small differences in the three equations are not readily detectable for this time period.

based on Eq. (5). However, the electrolyte would become depleted during such large time periods, and Eq. (5) would no longer apply.

The very small effects of changes in the cell heat capacity, as shown in Fig. 2, likely led Fleischmann to start using

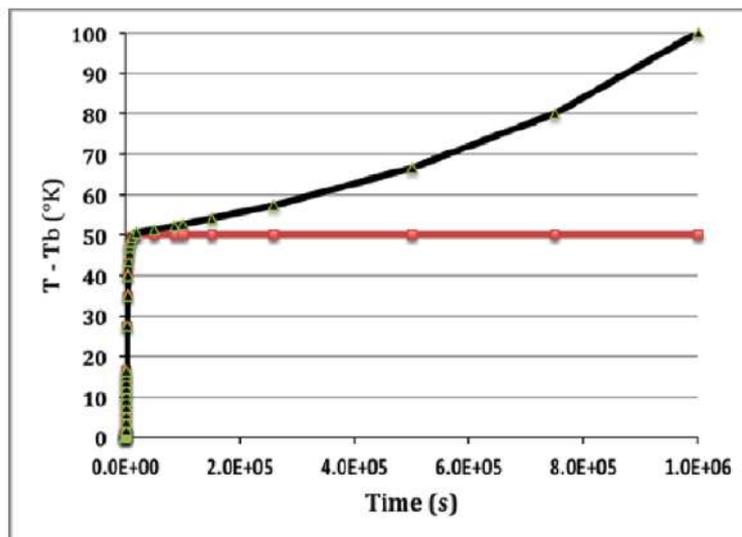


Figure 2. Cell temperature versus time for the first eleven days of cell operation based on Eqs. (3), (5) and (6). The black line gives the correct temperature-time behavior based on Eq. (6). The small differences between Eqs. (3) and (5) are not readily detectable for this time period.

the approximation of $C_p M d\Delta\theta/dt$ rather than the more exact term expressed by [15]

$$C_p d/dt(M\Delta\theta) = C_p M d\Delta\theta/dt + C_p \Delta\theta dM/dt, \quad (7)$$

where

$$M = M^\circ - (1 + \beta')It/2F \quad \text{and} \quad dM/dt = -(1 + \beta')I/2F. \quad (8)$$

Note that β' is a dimensionless term that accounts for the loss of water (H_2O or D_2O) due to evaporation or any causes other than electrolysis alone, and M is the equivalent moles of H_2O or D_2O that would give the total heat capacity of the calorimeter [15].

7. Summary

These excerpts from his letters show Martin Fleischmann's noble character and his scientific greatness. Fleischmann's mathematic skills are obvious from his modelling of the isoperibolic calorimetric system that should someday be recognized and applied to various electrochemical systems.

Acknowledgements

This work was supported from an anonymous fund at the Denver Foundation. An adjunct faculty position at the University of LaVerne is also acknowledged. The Author also thanks David L. Miles for the preparations of Figs. 1 and 2.

Appendix

The terms used by Fleischmann In Eq. (1) are defined as follows:

$$\alpha = Q_1/C_p M^\circ (\text{Ks}^{-1}) \quad \text{where} \quad Q_1 = (E - E_H)I + Q_f, \quad (A.1)$$

$$\beta = I/2FM^\circ \text{ s}^{-1}, \quad (A.2)$$

$$\gamma = (4k'_R\theta_b^3 + \psi I/\theta_b)/C_p M^\circ \text{ (s}^{-1}\text{)}, \quad \text{where} \quad \psi = E - E_H, \quad (A.3)$$

$$y = 2k'_R\theta_b^3(1 + \lambda)I/FC_p(M^\circ)^2 \text{ (s}^{-2}\text{)}, \quad \text{where} \quad \lambda = 0.0909, \quad (A.4)$$

$$\Delta\theta = T - T_b(\text{or } \theta - \theta_b)(\text{K}). \quad (A.5)$$

Typical values used for the Fleischmann–Pons Dewar calorimetry in 1992 are $\alpha = 1$ to $2 \times 10^{-2} \text{Ks}^{-1}$, $\beta = 1.0 \times 10^{-6} \text{s}^{-1}$ for $I = 0.80 \text{A}$, $\gamma = 4 \times 10^{-4} \text{s}^{-1}$, and $y = 2 \times 10^{-10} \text{s}^{-2}$. Most of these terms have been defined elsewhere [10,11,15].

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