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An Improved Rubidium Consumption Model for Discharge Lamps Used in Rubidium Frequency Standards

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I. INTRODUCTION

Rubidium (Rb) frequency standards typically employ Rb electrodeless discharge lamps to supply the light needed for optical pumping, a process central to the operation of the standard. The standard's reliability has been shown, in a number of instances, to be limited by the reliability of the discharge lamp (Ref. 2). In satellite applications, such as the global positioning system (GPS), standard reliability is of the utmost importance. Consequently, Rb discharge lamp reliability is of great concern. One goal of The Aerospace Corporation's atomic clock research program has been to ensure the reliability of the lamps employed in satellitecompatible Rb frequency standards.

A typical Rb discharge lamp is illustrated in Fig. 1. Composed of an alkali-resistant glass envelope, the lamp contains an initial Rb fill of hundreds of micrograms. An unlimited amount of Rb cannot be placed in the lamp because of concern over the stability of the lamp's output intensity. To allow the radio frequency (rf) discharge, several torr of a rare gas are also placed within the glass envelope. Lamp failure is indicated when the Rb resonance radiation required for optical pumping is no longer emitted by the discharge. In a previous publication (Ref. 1), a very detailed analysis of the causes of Rb lamp failure was presented. Briefly, the metallic Rb initially placed in the lamp was found to disappear during operation. The major metallic Rb loss mechanism is diffusion of Rb into the lamp's glass walls. A less significant loss resulted from an initial, and presumably, instantaneous, reaction between Rb and impurities within the lamp's envelope. The rate of Rb loss, or its consumption C, was well modeled by an equation of the form

$$C = A + B \sqrt{T}$$
(1)

where T is lamp operating time and A and B are empirical parameters determined by monitoring the loss of metallic Rb during lamp operation (Ref. 1).



Fig. 1. Schematic Drawing of a Rubidium Discharge Lamp. The internal surface areas of the Corning 1720 glass lamps studied are approximately 60% those of the Schott 8436 glass lamps investigated.

To extract A and B, consumption data vs operating time is fit to Eq. (1) using a least-squares procedure. The first term on the right-hand side of Eq. (1) reflects the rapid reactive loss of Rb, whereas the second term results from the more gradual diffusive loss mechanism.

Once A and B have been determined for a particular lamp design, Eq. (1) can be employed to predict Rb consumption for any period of lamp operation. Based on the consumption data available at the time of reference 1, Eq. (1) appeared to describe the consumption process very accurately. As more data have been obtained, it appears that the reactive loss mechanism is not truely instantaneous but occurs over a period of months. In this report, we analyze Rb consumption data treating A as a time-dependent quantity. We find that long-term consumption predictions then become less sensitive to the amount of data analyzed, and more accurate projections can often be made employing less data. Also, we find that when limited amounts of consumption data are used, Eq. (1) typically tends to overestimate long-term Rb consumption. Equation (1) is then conservative and still adequate for lamp reliability predictions.

In Section II, the observations that lead to a time-dependent A are discussed. The statistics of the modified consumption model are developed in Section III and applied to experimental data in Section IV. Conclusions are presented in Section V.

II. OBSERVATIONS THAT LED TO A RECONSIDERATION OF THE CONSUMPTION EQUATION

Many of the Rb discharge lamps being tested at Aerospace have been operated continuously for several years, with interruptions only for the acquisition of consumption data. The method for acquiring the consumption data has been previously described (Ref. 1), and, for the purposes of this discussion, is not important. As data were accumulated and subsequently fit to Eq. (1), certain trends became evident. The value for the intercept A obtained from the fitting procedure appeared to be decreasing monotonically. In addition, this decrease over time appeared to be asymptotic with respect to some final values for the intercept A and the slope B. Table 1 lists the intercepts and slopes of Eq. (1) for consumption data from four GPS-style Schott 8436 lamps produced by Efratom, a Division of Ball Corporation, and six Corning 1720 lamps produced by EG&G, Electronic Components Division. For the EG&G lamps, A appears to be rising to some value around 19 or 20 μ g, whereas B is falling to a value near 0.15 $\mu g/\sqrt{h}$. For the Schott lamps, it appears possible that A may eventually surpass 20 μ g, and B may fall below 0.4 μ g/ \sqrt{h} . Note that the changes are generally smaller as more data are included into the fit, for both Corning 1720 and Schott 8436 lamp data. Thus the A's and B's appear to be asymptotically approaching some final values.

One way to readjust Eq. (1) to reflect the observed trends is to introduce a time dependence for both A and B, so that Rb consumption C = $A(T) + B(T) \sqrt{T}$. However, the model equation is directly related to simple diffusion theory, in which M, the amount of material consumed, is given by $M = 2AC_0 \sqrt{(\pi/D)}$, where A is the surface area, C_0 is the density of the penetrating species at the glass surface, and D is the diffusion coefficient (Ref. 3). B, in principle, is equal to $2AC_0 \sqrt{(\pi/D)}$. Neither the surface area nor the diffusion coefficient has a clear time dependence. The density of the penetrating species, Rb metal in the vapor phase, should remain constant at the glass surface, as long as there is Rb

For data taken to: (yr)	Α (µg)	B (µg/√h)
0.80	-2.6±5.9	0.864±.122
1.14	-5.0±5.6	0.930±.100
1.71	-3.8±5.5	0.902±.086
2.28	8.4±6.1	0.638±.081
3.43	12.8±5.9	0.550±.069
4.00	15.2±5.5	0.506±.057
4.57	18.1±5.4	0.456±.050

Table 1. Consumption Parameters, A and B, as Functions of Increasing Periods of Lamp Operation

EG&G Corning 1720 Glass Lamps

Efratom Schott 8436 Glass Lamps

For data taken to: (jr)	A (μg)	B (μg/√h)
0.29	13.3±2.9	0.341±.093
0.46	14.1±2.5	0.304±.069
1.37	15.3±2.0	0.261±.047
2.06	16.7±1.7	0.181±.030
2.28	18.3±1.4	0.176±.020
2.85	18.2±1.4	0.181±.019
3.20	18.7±1.3	0.168±.016

metal left in the liquid reservoir of the lamp to constantly supply the vapor. Only at failure, when all the Rb metal in the liquid reservoir is gone, would the constancy of the penetrating species not be met. It would not be consistent with this model to assume a time dependence for B.

In contrast to B, it is reasonable that A would have some time dependence. A represents the amount of metallic Rb lost through a reaction of the form

$$Rb + I \xrightarrow{k} RbI$$
 (2)

where I represents impurities within the lamp at its fabrication, RbI is a nonvolatile reaction product, and k is the reaction rate constant. While this reaction was initially assumed to be instantaneous, this is not necessarily the case. With the concentration of Rb within the lamp constant, the time dependence of the amount of I is given by

$$[I] = [I]_{o} e^{-DT}$$
(3)

with

$$D = k [Rb]$$

where the brackets imply the amount of a particular species within the lamp and the subscript zero indicates initial amount. The time dependence of the amount of metallic Rb lost through Eq. (2) is then given by

$$A(t) = A' (1 - e^{-DT})$$
 (4)

where A' represents the total amount of Rb lost through reaction. The expanded consumption equation is

$$C = A'(1 - e^{-DT}) + B'\sqrt{T}$$
(5)

Subsequently, this equation will be used to analyze consumption data initially studied using Eq. (1). We now need to introduce the statistical procedures by which the parameters and the performances of Eqs. (1) and (5) can be compared.

III. CONSUMPTION STATISTICS

Practical lamp reliability analyses use Rb consumption data obtained over limited periods of lamp operation, in order to predict long-term consumption. We wish to obtain most-probable Rb consumptions and their variances for any operating times. Equation (1), which may be put in a linear form, can be analyzed using normal linear least-squares techniques. However, Eq. (5) cannot be put in linear form and requires a more sophisticated analysis. We employ a general approach applicable to both equations.

Let C_i be a consumption measurement made after a lamp operating time t_i . Let p be the number of such measurements made. The functional form to which the measurements will be fit is $\overline{C}(t) = f(a_k, t)$, where a_k represents the parameters of interest. Assuming the measurements, C_i , are Gaussian distributed with standard deviations σ_i , a likelihood function, L, may be written⁴,⁵

$$L = \prod_{i=1}^{p} \frac{1}{\sqrt{2\pi} \sigma_{i}} \exp \{-[C_{i} - \overline{C}(t_{i})]^{2}/2\sigma_{i}^{2}\}$$
(6)

Setting W = ln(L), we find

$$W = -\frac{1}{2}M - \sum_{i=1}^{p} \ln \left(\sqrt{2\pi} \sigma_{i}\right)$$

with

$$M = \sum_{i=1}^{p} [C_{i} - \overline{C}(t_{i})]^{2} / \sigma_{i}^{2}$$
(7)

The "best" parameter values, a_{ν}^{*} , are solutions of the set of equations

$$\frac{\partial M}{\partial a_k} = 0 \tag{8}$$

The least-squares variances for the a_k^{\bullet} are given by

$$\overline{(a_{\ell} - a_{\ell}^{*})(a_{m} - a_{m}^{*})} = (H^{-1})_{\ell m}$$
(9)

where the bar implies averaging over many sets of consumption measurements, each yielding a set of parameters, a_k . $(H^{-1})_{lm}$ is the l-th row, m-th column element of the k-row by k-column matrix, whose inverse has elements defined by

$$H_{rs} = \frac{1}{2} \frac{\partial^2 M}{\partial a_r \partial a_s}$$
(10)

Another quantity of interest is the variance for a predicted Rb consumption at a time t_p . The predicted consumption is $\overline{C}^{*}(t_p)$, equal to $f(a_{k}^{*}, t_p)$, while its variance at t_p is specified by^{4,5}

$${}^{\sigma}\overline{C}^{*}(t_{p}) = \sum_{\mathfrak{L}} \sum_{\mathfrak{m}} \frac{\partial \overline{C}(t_{p})}{\partial a_{\mathfrak{L}}} \frac{\partial \overline{C}(t_{p})}{\partial a_{\mathfrak{m}}} (H^{-1})_{\mathfrak{L}\mathfrak{m}}$$
(11)

The quantity that is of most interest in making a lamp life prediction is the Rb consumption C_p at t_p , specified with a certain confidence. Based on the normal distribution, a 90% confidence that a lamp will consume no more than C_p results if

$$C_{p} = \overline{C}^{*}(t_{p}) + 1.282 \sigma_{\underline{T}}^{*}(t_{p})$$
 (12)

All of the analysis done in this report is on the collective body of consumption data for a particular type of lamp. That is, all consumption data for GPS-style Schott 8436 lamps are lumped into one data set, as are all the consumption data for the Corning 1720 lamps. While it is possible to analyze each lamp individually, we have chosen to maintain consistency with the previous analysis (Ref. 1).

IV. APPLICATION OF RUBIDIUM CONSUMPTION FITTING TECHNIQUES TO LAMP DATA

The EG&G 1720 glass lamp consumption data were analyzed using the techniques presented in Section III. In Table 2, the parameter values for Eqs. (1) and (5) for various lengths of analyzed data are presented. Additionally, the most probable and the 90% confidence level consumptions have also been projected for 11 years of operation. While A and B display clear trends as lamp operating times are increased, no such trends are apparent for A', D, and B'. This gives us confidence in our supposition of a time-dependent reactive consumption mechanism. The most probable 11-year consumptions also show similar behaviors. For these lamps, Eq. (5), using data based only on one-half of a year's lamp operation, yields accurate long-term projected consumption. For Eq. (1) to yield similar mostprobable consumptions, 2 years of lamp operation are required. In Fig. 2, fits to 0.29 and 3.20 years of consumption data for both equations are presented. As expected, fits resulting from Eq. (5) do not change significantly between the two operating times, whereas Eq. (1) fits show clear variations. The knee in the Eq. (5) fits, occurring at about 500 to 1000 h of operation, results from the completion of reactive consumption and growth of the diffusive process. In terms of 90% confidence limits, both equations yield similar predictions. The reason for this apparent contradiction is that the uncertainties associated with the Eq. (5)analysis are typically greater than those resulting from an analysis based on Eq. (1). If the additional parameter in Eq. (5) is included, more data are needed to obtain consumption predictions with levels of confidence similar to those resulting from Eq. (1).

The same analyses were applied to the Schott 8436 glass lamp consumption data. The results are summarized in Table 3. The trends displayed by the parameters associated with Eq. (1) are the same as the trends displayed by the Corning 1720 glass lamps. Using Eq. (5), meaningful fits did not result until after 4 years of data had been analyzed. The reason is

Data
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Glass
1720
Corning
of
Analysis
Table 2.

Lamp Data Analyzed (yr)	A (ng)	A B_ (ug) (ug//h)	C*(11 yr) (ug)	C*(11 yr) C _p (11 yr) (ug) (ug)	А' (µg)	(10 ⁻³ /h)	^B - (μg/Λh)	Č#1(11 yr) (ug)	C*'(11 yr) C' (11 yr) (ug) P(ug)
0.29	13.3±2.9	13.3±2.9 0.341±.093	119±26	153	21.5±9.6	21.5±9.6 4.8±3.0	0.137±.234	64±63	145
0.46	14.1±2.5	14.1±2.5 0.304±.069	109±19	133	20.3±6.4	5.2±2.9	0.168±.144	73±38	122
1.37	15.3±2.0	15.3±2.0 0.261±.047	96±13	113	19.5±3.7	5.6±2.5	0.184±.073	77±19	101
2.06	16.7±1.7	16.7±1.7 0.181±.030	73±8	83	19.6±2.3	5.5±2.0	0.181±.035	76±9	87
2.28	18.3±1.4	18.3±1.4 0.176±.020	73±5	80	21.6±2.0	4.7±1.4	0.142±.024	9 2 99	73
2.85	18.2±1.4	18.2±1.4 0.181±.019	75±5	81	21.1±1.9	4.8±1.5	0.151±.022	68±5	75
3.20	18.7±1.3	18.7±1.3 0.168±.016	71±4	76	21.8±1.8	4.6±1.4	0.140±.020	65±5	71



Fig. 2. Most-Probable Rubidium Consumptions vs Lamp Operating Time for Corning 1720 Glass Lamps Based on the Standard Model, Eq. (1) (dashed line), and the Expanded Model, Eq. (5) (solid line). Years of consumption data used in fitting procedures: (a) 0.29, (b)3.20.

Table 3. Analysis of Schott 8436 Glass Lamps' Consumption Data

Lamp Data Analyzed (yr)	А (уц)	А В (иg) (иg//ћ)	C*(11 yr) (μg)	C*(11 yr) C _p (11 yr) (μg) (μg)	А' (иg)	^{D4} /h) (μg//h)	В' (нg//h)	C**((1: yr) (µg)	C#'(1: yr) C ['] (11 yr) (ug) ^p (ug)
2.28	8.4±6.1	8.4±6.1 0.638±.081	206±20	232	I	I	ı	ı	ł
3.43	12.8±5.9	12.8±5.9 0.550±.069	184±17	205	ı	ı	ı	ı	ı
4.00	15.2±5.5	15.2±5.5 0.506±.057	172±13	189	73.3±22.0	2.9±.8	0.100±.149	104±25	137
4.57	18.1±5.4	18.1±5.4 0.456±.050	160±11	174	71.1±18.7 2.9±.9	2.9±.9	0.117±.118	107±19	132

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apparent when Fig. 3, which displays all consumption data fits to both functional forms, is reviewed. The knee region in the Eq. (5) fit has moved out to between $10 - 40 \times 10^3$ h of operation. This fit would indicate that, only after about 5×10^4 h of operation, nearly 5 years, does the long-term diffusive process become significant. At times less than this, consumption is dominated by the reactive component, and attempts to extract diffusive parameters are not successful. In Table 3, we see that the D coefficient is only about 6% the size of that in Table 2, consistent with the apparently slow nature of the reactive process for this glass.

After 3 years of operation of the Corning 1720 glass lamps, B and B' were very close in magnitude. After 4.7 years of operation of the Schott 8436 lamps, B and B' are still quite different. While B continues to decrease, it may require many more years before B actually matches B'. This is important, as the two model equations yield significantly different relative consumption contributions from reactive and diffusive processes. As the extremely long-term consumption will be dominated by diffusion, when the merits of various glass types are compared, it is desirable to have accurate diffusive parameters. One way to determine the relative contributions of diffusive and reactive consumption in Schott 8436 glass is to perform simple chemical analyses on the lamp envelopes. As we believe the reaction product resides primarily on the envelope's inner surface, washing this surface with water and comparing the Rb content of the rinsings with the Rb remaining in the glass would be useful. However, we do not intend to do this, because these lamps are valuable long-term test subjects. We lean toward the diffusive coefficient produced by Eq. (5) as most representative of Schott 8436, and hence feel, considering long-term Rb consumption, that it is an excellent glass, probably superior to Corning 1720.



Fig. 3. Most-Probable Rubidium Consumption vs Lamp Operating Time for Schott 8436 Glass Lamps Based on the Standard Model, Eq. (1) (dashed line), and the Expanded Model, Eq. (5) (solid line). A total of 4.57 years of consumption data were used in fitting procedures.

V. CONCLUSIONS

We have observed that the parameters resulting from fitting Rb consumption data to the standard consumption model $C = A + B \sqrt{T}$ often depend upon the length of lamp operating time spanned by the data. Typically, as operating time increases, the value of A increases, while the value of B decreases. This dependence is attributed to a nonconstant A. When A is modified to include a simple time dependence, resulting from first-order chemical reaction kinetics, $\{A(T) = A'[1 - exp(-DT)]\}$, the parameters of the expanded model no longer display this sensitivity to the length of lamp operation.

The underlying importance of fitting the consumption data to one model or another is the reliability ascribed to a lamp based on the fit used. A consistent consumption prediction enhances the respect one has for a particular model. On the other hand, one can argue for the simpler model, particularly as we know that its predictions will, in general, be overly pessimistic. Furthermore, when consumption data obtained at short operating times are fit to the expanded model, a larger standard deviation is associated with the 11-year consumption prediction. This translates into an insignificant difference in the 11-year consumption number, with a 90% level of confidence. It is clear, from Table 3, that the expanded model needs more data than the simpler model to make consumption predictions with equal precision. This is because data must be fit to an equation with three parameters instead of just two. Because the consumption predictions from the expanded model are so consistent, regardless of the additional complications of fitting to an equation with three parameters, we believe that the expanded model makes more accurate consumption predictions that are based on a more physically correct model. Also, we feel that the expanded model allows more accurate assessment of a glass's long-term diffusive behavior. At this point, though, we don't recommend discarding the standard model. Because of its simplicity and conservative approach to lamp reliability, the standard

model remains quite attractive. As additional long-term consumption data are generated, a more definitive statement will be possible.

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<u>Electronics Research Laboratory</u>: Microelectronics, solid-state device physics, compound semiconductors, radiation hardening; electro-optics, quantum electronics, solid-state lasers, optical propagation and communications; microwave semiconductor devices, microwave/millimeter wave measurements, diagnostics and radiometry, microwave/millimeter wave thermionic devices; atomic time and frequency standards; antennas, rf systems, electromagnetic propagation phenomena, space communication systems.

<u>Materials Sciences Laboratory</u>: Development of new materials: metals, alloys, ceramics, polymers and their composites, and new forms of carbon; nondestructive evaluation, component failure analysis and reliability; fracture mechanics and stress corrosion; analysis and evaluation of materials at cryogenic and elevated temperatures as well as in space and enemy-induced environments.

<u>Space Sciences Laboratory</u>: Magnetospheric, auroral and cosmic ray physics, wave-particle interactions, magnetospheric plasma waves; atmospheric and ionospheric physics, density and composition of the upper atmosphere, remote sensing using atmospheric radiation; solar physics, infrared astronomy, infrared signature analysis; effects of solar activity, magnetic storms and nuclear explosions on the earth's atmosphere, ionosphere and magnetosphere; effects of electromagnetic and particulate radiations on space systems; space instrumentation.

