GAS EVOLUTION, DRYOUT, AND LIFETIME OF VRLA CELLS - AN ATTEMN' TO CLARIFY FIFTEEN YEARS OF CONFUSION AND MISUNDERSTANDING

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ABSTRACT:

This paper compares a reasonable sample of the more than 15 years published data on VRLA batteries in a single standardized fashion. Using the standardized criterion of ml of hydrogen per day per 1OOAH and calculated time to 10% water loss (which we have previously reported' is equivalent to capacity loss below 80% of rated value), we find:

Times to 10% water loss ranging overall from 6 to 1300 years from literature data on laboratory and field studies of positive grid corrosion, cell weight loss and actual quantities of gas collected for both AGM and GEL cells.

Laboratory corrosion data generally predict the longest times to 10% water loss (30-100 years), while positive grid corrosion in actual VRLA cells generally predict less than 10 years.

Direct gas collection results tend to predict greater lifetimes than field corrosion data, but are contradicted by the shorter lives predicted from weight loss measurements.

The requirements of various national and international VRLA Standards are also compared in this standardized fashion.

The extraordinary range and disparity of literature results, made obvious by the use of this standardized approach, suggests its more widespread application might be helpful in developing a clearer understanding of the mechanisms which govern VRLA performance, life and failure modes.

BACKGROUND:

Since the earliest development and introduction of recombinant VRLA cells, most investigators, in considering the overall material balance within the cell, in float application, have agreed on several principles:

To operate effectively, for long life, the recombinant cell must remain leak-free.

In a leak-free system, the overall net reactions, on float, will be dominated by the oxygen recombination cycle:

1. Oxygen generation at the positive electrode
2H20 = 02 + 4H+ + 4e-

2. Oxygen reduction at the negative electrode
02 + 4H+ + 4e- = 2H20

3. If recombination were 100% efficient, the above reactions would be sufficient to describe the overall float operation of the system.

4. However, since positive grid corrosion Pb + 2H20 = PbO2 + 4H+ + 4e-
cannot be prevented, under float charge conditions, electron balance requires corresponding balance at the negative plate.
4H+ + 4e- = 2H2

Thus, in their simplest form, these secondary reactions reduce oxygen recombination efficiency(ORE) below 100%, and must result in the emission of gase~us hydrogen, loss of oxygen to positive grid corrosion and, hence, permanent loss of water from the limited electrolyte within the cell.

In addition, there are other competing reactions:

5. Oxidation of carbonaceous materials to form carbon dioxide:

$C + 2H_2O = CO_2 + 4H^+ + 4e^-$

NOTE: Just as with lead corrosion at the positive grid, this reaction must also be balanced by hydrogen evolution at the negative:

$4H^+ + 4e^- = 2H_2$

6. Negative self discharge:

$Pb + H_2SO_4 = PbSO_4 + H_2$

Both of these reactions also reduce the overall oxygen or hydrogen in the system and hence lead to water loss.

The key point is that any reduction in oxygen recombination efficiency consumes water and results in hydrogen evolution.

Over the years, authors have published various values of predicted or measured rates of positive grid corrosion. Likewise, rates of negative plate self discharge have been published and estimates have been made of the oxidation of carbonaceous materials. There have been attempts to collect and measure the actual amount of hydrogen evolved from VRLA cells, both by volumetric collection or weight loss techniques, while on extended float service. Consequently, in considering the possible effects of water loss, or dryout, on the life expectancy of VRLA cells, it would seem reasonable to turn to the published literature for guidance and attempt to relate these published values to specific designs and conditions of use. Unfortunately, these values, whether calculated or measured, have typically been presented in a variety of units, so that direct comparison of all of these results has been made difficult and if not impossible, certainly, non-obvious.

It is the purpose of this paper to review a significant portion of the relevant published work, but then to convert all the data into one common set of units, to facilitate direct, side by side comparisons. To do this, the authors have adopted a recently published approach, which relies on a simple, easily understood concept.

At INTELEC '95, Jones and Feder introduced the following combination of concepts[1]

Based on both published and unpublished data, a loss of 10% of the water from a VRLA cell would reduce its capacity to below 80% of its initial, rated value[2,3,4]

Therefore, if a cell were to last 20 years, this 10% loss can easily be translated into a maximum allowable rate of hydrogen evolution and normalized to a standardized cell size. In their work, this allowable rate was calculated to be no greater than 20 ml of hydrogen per day per 1OOAH of cell capacity.*

*While some may question the universal validity of this value, it will serve as a constant to which all data may be compared.

INTRODUCTION

This paper examines both early and recently published information, both measured in the laboratory or in actual cell use, on the rate of positive grid corrosion, expressed either as corrosion current or PbO_2 penetration, published gas collection or weight loss results and includes test information and criteria contained in both national and international VRLA standards. All these data are converted into the standardized unit of ml of hydrogen per day per 1OOAH of cell capacity. They are then compared to the 20 year target of 20 ml of hydrogen per day per 1OOAH of cell capacity. From this comparison, predicted life to 10% water loss/capacity loss is calculated.

Once a single standard unit has been established, it is possible to compare the resulting range of values and the range of predicted lifetimes.

It is the intent of this paper to document this wide range of values, primarily to call attention to the problems associated with the variable performance characteristics of VRLA batteries, the consequent difficulties of obtaining meaningful test results which have generic applicability and perhaps, most of all, to highlight our continuing lack of understanding of the interacting mechanical, chemical and electrochemical performance intricacies which govern their behavior, aging and ultimate failure.

Hopefully, results of these comparisons may help to focus the dialogue on the true mechanisms and performance capabilities of VRLA cells used in stationary float service.

LITERATURE RESULTS:

1. Positive Grid Corrosion

1.1 Laboratory Studies

Starting with the classic work of Lander6, many papers have appeared which provided quantitative values of corrosion of lead or lead alloys, under conditions which simulated their usage as positive grids of lead acid batteries in float service. It is not the intent of this paper (nor would it be possible) to present and compare all of these published values. Instead, we have selected some of the most widely quoted, under conditions which seem applicable to VRLA batteries used in Telecommunications continuous float operation.

The results are shown in Table I, which asumes a positive grid surface area of 1174 sq. cm. per 1OOAH. These results immediately highlight the rationale for this paper. Starting with Lander's earliest work on the corrosion of pure lead, corrosion rates are shown in mg/34 sq. cm/hr for exposure times of only 24 hours or less and at positive polarizations ranging from 25 to 225 mv above open circuit values. Five years later, Lander reported data for much longer exposure times (140 hours) and reported either weight loss (mg./sq.cm./140 hr) or penetration rates in cm/year. His results, normalized as described above, range from 6 ml/day/1OOAH to 58 ml/day/1OOAH under generally comparable conditions of usage.

Table I also includes results of laboratory studies of PbCa alloys, both by Lander7 subsequent data on pure Pb or PbCa by Ruetschi8, Willenganz10, Milner (and Baker)9 and Fiorino (and Baker)11. In normalized units, for VRLA designs, the range of these values now extends from 1.1 ml/day/100AH to 64 ml/day/100AH. For these laboratory studies alone, the values project to times to 10% water loss ranging from 6.3 to 364 years!

1.2 Field Service Results

Table II shows measurements of positive grid corrosion from VRLA cells in actual use , whether in laboratory float studies or actual field usage. TIme periods ranging from approximately two months to 12 years are reported (primarily in INTELEC) in publications from 1982 to 1995.

From these results, one must conclude that positive grid corrosion in actual cells is significantly laboratory. Here the normalized hydrogen evolution results range from 11 ml/day/100AH, which convert to times ranging from 4 years to 36 years to 10% water loss. Most of the results suggest less than ten (10) years

TABLE I

TIME TO 10% WATER LOSS
LABORATORY GRID ALLOY CORROSION MEASUREMENTS

DATE	REF	ALLOY	POS. POL'N	TEMP °C	TEST PERIOD	MEASUREMENT	NORMALIZED ML/DAY/100AH*	YEARS TO 10% WATER LOSS
1951	5	Pb	25mv	30°C	24Hr	0.186mg/34cm²/Hr	36.6	10.9
1951	5	Pb	125mv	30°C	16Hr	0.297mg/34cm²/Hr	57.8	6.9
1951	5	Pb	225mv	30°C	24Hr	0.225mg/34cm²/Hr	44	9.1
1956	6	Pb	200mv	27°C	140Hr	0.125mg/cm²/140Hr	5.9	68
1956	6	Pb	200mv	30°C	140Hr?	0.0009cm/yr	7.5	53
1956	6	Pb	100mv	30°C	140Hr	0.0012cm/yr	10.1	39
1956	6	Pb	?	30°C	>140Hr	.003cm/yr	25.2	16
1958	7	PbCa	75mv	25°C	?	0.56mils/yr	11.9	34
1964	8	PbCa (.075)	100mv	30°C	24Hr	5ua/cm²	64	6.3
1970	9	PbCa	70mv	25-30°C	>100 Hr	0.2uA/cm²	2.2	182
1979	10	PbCa	70mv?	25-30°C	10 yrs	3 mils	6.4	63
1984	11	Pb	80mv	30°C	>100Hr	4.9uA/cm²	63	6.3
1984	11	Pb	80mv	30°C	>100Hr	0.1uA/cm²	1.1	364

*NOTE: CALCULATION ASSUMES 1174 cm² SURFACE AREA/100 AH GRID

TABLE II

TIME TO 10% WATER LOSS POSITIVE GRID CORROSION MEASUREMENTS								
DATE	REF	CELL TYPE	CELL SIZE BATT'Y SIZE	FLOAT VOLTAGE	TEST PERIOD	MEASUREMENT REPORTED	NORMALIZED ML/DAY/100AH*	YEARS TO 10% WATER LOSS
1982	12	AGM	100AH	2.25	64 DAYS	3.0mils/yr	64	6.3
1988	13	AGM	500AH?	2.25	19 MONTHS	2.13mils/yr	45	8.9
1988	13	AGM	500AH?	2.30	19 MONTHS	4.77 mils/yr	102	3.9
1988	13	AGM	500AH?	2.25@43°C	19 MONTHS	2.85 mils/yr @ 43°C	61 @ 43°C	6.6 @ 43°C
1993	14	--	CORROS. TEST	2.25	NOT STATED	0.05mm/yr	42	9.5
1993	14	AGM	NOT STATED	2.25	12 YEARS	0.025mm/yr	21	19
1994	15	AGM	600 AH	NOT STATED	19 MONTHS	3 mils/yr	64	6.3
1994	16	AGM	NOT STATED	NOT STATED	NOT STATED	0.02mm/yr to 0.04mm/yr	17 to 34	24 to 12
1995	17	AGM	500-1000AH	2.25	6 - 7 YEARS	0.5 mils/yr to 1.8 mils/yr	11 to 38	36 to 11
1995	18	AGM	800 AH	2.25	6 YEARS	0.8 mils/yr to 4.4 mils/yr	14 to 94	29 to 4.3
1995	19	AGM	550 AH	2.25	3 YEARS	20 u to 40 u	17 to 34	24 to 12

* NOTE: CALCULATION ASSUMES 1174 cm^2 SURFACE AREA/100 AH GRID

usage to 10% water loss.

2. Weight Loss Measurements

Since the introduction of VRLA cells various workers have attempted to measure gas loss (hydrogen) or perhaps water loss, directly by loss of weight. This method was generally popular in early VRLA publications, but posed significant experimental difficulties, both in obtaining sufficient gravimetric sensitivity and in being assured that the weight loss was due solely to hydrogen and not a combination of gasses, including water vapor itself.

Table III shows results of a few such measurements, published over the peroid form 1984 to 199420,21,22. Again, the results are presented in a variety of units , for a variety of cell and battery sizes. However, once normalized, they range from 4ml/day/100AH to 31 ml/day/100AH. These values covert into times ranging from 6.5 to 10% water loss.

TABLE III

TIME TO 10% WATER LOSS WEIGHT LOSS MEASUREMENT								
DATE	REF	CELL TYPE	CELL SIZE BATT'Y SIZE	FLOAT VOLTAGE	TEST PERIOD	MEASUREMENT REPORTED	NORMALIZED ML/DAY/100AH	YEARS TO 10% WATER LOSS
1984	20	AGM	100AH 48V	2.27	12 WEEKS	0.77 gm	4.3	93
1989	21	AGM	?AH 168 MONOBLOCS	NOT STATED	4 YRS	80mg/AH/CELL	61	6.6
1994	22	AGM	50AH 2-12V	2.27	4 YRS	41 gm	52	7.6
1994	22	AGM	50AH 2-12V	2.23	4 YRS	42 gm	54	7.5
1994	22	AGM	50AH 2-12V	2.31	4 YRS	48 gm	61	6.5

3. Gas Collection Measurements

Considering the normalizing units chosen, ml of hydrogen/day/1OOAH, the direct collection of gases evolved on float should provide the most accurate and most meaningful results. However, all workers performing these measurements or reviewing their results, have repeatedly cautioned of the extreme sensitivity, both to obvious or minute leaks in the gas collection system, as well as the more subtle, but still significant loss of hydrogen by permeation through plastic collection tubing. A few workers have recommended exclusive use of metal collection tubing, but we have found only one published result in the literature27. Likewise, workers reporting gas collection from 1984 to 1994 have used a variety of cell sizes, battery configurations and collection times ranging from four (4) days to two (2) years. Here there was sufficient published data to justify separation into two tables. Table IV reflects information only on AGM cells, while Table V presents data on Gel designs.

3.1 AGM Cells

Looking first at Table IV for AGM designs, one is struck immediately by the variety of cell sizes, configurations and collection times involved and again by the fact that none of the data is easily compared. This becomes bl4tantly obvious, once the data are normalized, ranging from 0.3 to 28 ml/day/1OOAH. Translated into time to 10% water loss, the results range from 14 years to 1333 years! The latter value is even more interesting, since it was reported in the same paper in which measured positive grid corrosion suggested times to 10% water loss ranging from 4 to 9 years'3. Had these data all been normalized, the discrepancy would have been obvious. Likewise, the data from reference 20 shows 4.3 ml/day/1OOAH (93 years to 10% water loss). This gas emission was confirmed to be predominantly hydrogen, presumably due to positive grid corrosion of what were probably prototype battery designs. Field data representing the same type of cells, have subsequently been reported26 to have lost capacity and failed in 5 to 6 years. Post morta determined that the failures were due to excessive positive grid corrosion, which certainly would not have been expected from the gas collection data.

3.2 Gel Cells

Table V shows gas collection data reported for Gel cells, by several authors from 1984 to 1993, for both new and aged cells and over a range of float voltages from 2.23 volts per cell to 2.35 volts per cell. Again the normalized hydrogen evolution values extend from 10 ml/day/1 OOAH to 625 ml/day/1OOAH. Corresponding times to 10% water loss range from 0.6 to 40 years. The data provide an excellent example of the significant effect of float voltage and aging of the Gel on gas evolution.

TABLE IV

TIME TO 10% WATER LOSS GAS COLLECTION MEASUREMENT								
DATE	REF	CELL TYPE	CELL SIZE BATTERY SIZE	FLOAT VOLTAGE	TEST PERIOD	MEASUREMENT REPORTED	NORMALIZED ML/DAY/100AH	YEARS TO 10% WATER LOSS
1984	20	AGM	100 AH 48V	2.27	12 WKS	8601 ML	4.3	93
1984	20	AGM	100 AH 48V	2.40	4 WKS	9340 ML	28	14
1987	23	AGM	50 AH 12V	2.27 @ 40°C	20 WKS	24ML/AH/CELL @ 40°C	17 @ 40°C	23 @40°C
1988	13	AGM	600 AH	2.25	430 DAYS	0.07 ML/HR	0.3	1333
1989	21	AGM	300 AH 48V	2.27	2 YRS	11,000 ML/CELL	5.0	80
1994	15	AGM	600 AH	NOT STATED	NOT STATED	60 ML/DAY	10	40

TABLE V

TIME TO 10% WATER LOSS GAS COLLECTION MEASUREMENTS								
DATE	REF	CELL TYPE	CELL SIZE BATTERY SIZE	FLOAT VOLTAGE	TEST PERIOD	MEASUREMENT REPORTED	NORMALIZED ML/DAY/100AH	YEARS TO 10% WATER LOSS
1984	24	GEL	NOT STATED	2.30	NEW	1 ML/AH/DAY	100	4
1984	24	GEL	NOT STATED	2.30	3 YRS	0.1 ML/AH/DAY	10	40
1992	25	GEL	NOT STATED	2.30	4 DAYS	25ML/AH	625	0.6
1993	4	GEL	250 AH	2.35	NEW 4 DAYS	3.87 LITERS/ 4 DAYS	387	1.0
1993	4	GEL	250 AH	2.35	4 DAYS AFTER 400 DAYS	0.7 LITERS/ 4 DAYS	70	5.7
1993	4	GEL	NOT STATED	2.23	NOT STATED	0.1ML/AH/DAY	10	40
1993	4	GEL	NOT STATED	2.27	NOT STATED	0.6ML/AH/DAY	60	6.7
1993	4	GEL	NOT STATED	2.30	NOT STATED	1.4ML/AH/DAY	140	2.9

4. National and International Standards

Most national and international VRLA standards present requirements and test methods for determining the rate of gas evolution from VRLA cells on float and on overcharge. Table VI shows four such examples. All but the IEC show a requirement of a minimum recombination efficiency, ranging from 95% in the British and French standards to 99% (or 97%) in the US Telecommunication standard (The 99% vs 97% values have not been resolved by the US Tl/E1 .5 group as of this writing). Since one cannot calculate a rate of gas evolution without knowing the float current, we have shown no entries in the normalized ml/day/1OOAH column, for any of the recombination efficiency values. For the IEC standard, it is possible to calculate a normalized value, 100 ml/day/1OOAH at the maximum allowable rate under normal float conditions. This translates to 4 years life to 10% water loss.

To demonstrate the effect of float current in determining the rate of gas evolution at constant value of recombination efficiency, we have presented Table VII. This table reproduces data shown by the authors at the 1995 INTELEC Conference' on actual measured float currents on three pairs of cells, three different manufacturers and three different ampere hour capacities. Actual gas collection data were reported for these cells and are shown on the table and compared to the calculated values.
The table shows the three float currents, for the three sets of cells, as measured and as normalized to ma/1OOAH, when the cells were on float at 2.27 vpc. It then calculates the normalized gas evolution and times to 10% water loss for each float current and for 95%, 97% and 99% recombination efficiencies, as required by the various standards.

Note first that the float currents, when normalized, range from 44 to 160 ma/i OOAH for real cells measured under identical float conditions. The effects of the recombination efficiencies range from 17.5 to 87.5 ml/day/1OOAH as efficiencies decrease from 99% to 95% for the high float current cells. Overall, for the three sets of cells, at 95% recombination efficiency, gas evolution ranges from 24.1 to 87.5 ml/day/1OOAH. Time to 10% water loss range from 4.6 to 16.6 years. Hence a constant requirement for recombination efficiency is meaningless in predicting the amount of gas evolution, unless the float current is also stated.

As a matter of interest, note that the actual measured gas evolutions reported in the 1995 paper all fall at the lower values of recombination efficiencies, generally bracketing the 95% value.

TABLE VI

TIME TO 10% WATER LOSS VRLA BATTERY STANDARDS									
DATE	REF	CELL TYPE	CELL SIZE BATT'Y SIZE	FLOAT VOLTAGE	FLOAT CURRENT	TEST PERIOD	MEAS. REPORTED (REQUIREMENT)	NORMALIZED TO ML/DAY/100AH	YEARS TO 10% WATER LOSS
1987	BRIT.STD BS 6290 PART 4	AGM OR GEL	ANY	MFR'S RECOMM VALUE	EQUIL'B VALUE AT FLOAT VOLTAGE	180 DAYS	AH OF H_2 $\leq$ 5% OF TOTAL AH	VALUE DEPENDS ON FLOAT CURRENT*	VALUE DEPENDS ON FLOAT CURRENT*
?	FRENCH STD. VRLA NR 5641	AGM OR GEL	ANY	2.35	EQUIL'B VALUE @ 2.35V	400 DAYS	95% RECOMB. EFFIC. (ORE)	VALUE DEPENDS ON FLOAT CURRENT*	VALUE DEPENDS ON FLOAT CURRENT*
1996	US TELECOM T1E1.5 DRAFT VRLA STD.	AGM OR GEL	ANY	MFR'S RECOM. VALUE (UPPER LIMIT)	EQUILIB. VALUE AT FLOAT VOLTAGE	4 DAYS OR 100ML	97% OR 99% RECOMB. EFFIC. (ORE)	VALUE DEPENDS ON FLOAT CURRENT*	VALUE DEPENDS ON FLOAT CURRENT*
1993	IEC TC 82 STD 896-2	AGM OR GEL	12V STRING OF CELLS OR 24V SERIES OF MONOBLOC BATT'YS	MFR RECOM VALUE (UPPER LIMIT)	EQUILIB. VALUE AT FLOAT VOLTAGE	30 DAYS	$\leq$ 30 ml CELL-AH FOR 30 DAYS TOTAL	100	4

*SEE TABLE VII FOR EFFECT OF FLOAT CURRENT

TABLE VII

EFFECT OF FLOAT CURRENT ON TIME TO 10% WATER LOSS AT RECOMBINATION EFFICIENCIES FROM 95 - 99%						
CELL TYPE	CELL SIZE	I_{float} @ 2.27V_f ma	I_{float} ma/100AH	ORE%	NORMALIZED ML/DAY/100AH	YEARS TO 10% WATER LOSS
AGM	125AH*	200*	160*	95%	87.5	4.6
			160	97%	52.5	7.6
			160	99%	17.5	22.9
			160		AS MEASURED* 64 - 112	6.3 - 3.6
AGM	180 AH*	80*	44	95%	24.1	16.6
			44	97%	14.5	27.6
			44	99%	4.8	83.3
			44		AS MEASURED* 28 - 44	14.3 - 9.1
AGM	140 AH*	160*	114	95%	62.5	6.4
			114	97%	37.5	10.7
			114	99%	12.5	32
			114		AS MEASURED* 42.9 - 100	9.3 - 4

* ACTUAL VALUES FOR 3 PAIR OF CELLS
REPORTED BY JONES & FEDER, INTELEC 1995

SUMMARY & CONCLUSIONS

Uncertainties and contradictory information regarding the life, performance and failure modes of VRLA cells have become increasingly apparent from papers presented at INTELEC and other conferences over the last five to six years. Perhaps even more significant are the vigorous discussions attempting to explain the "true" technical mechanisms which govern VRLA behavior.

The purpose of this paper has been, quite simply, to compare a reasonable sample of the published data on VRLA batteries in a single, standardized fashion. The results, we believe, directly highlight the wide range and often contradictory values obtained, in a clear, easily understood and unequivocal fashion. At the very least, they also raise serious questions as to our understanding of the mechanisms which truly govern VRLA performance, life and failure modes.

Using the standardized criteria of ml of hydrogen per day per 1OOAH and calculated time to 10% water loss (which we believe is equivalent to capacity loss to below 80% of rated value), we find:

1. Laboratory grid alloy corrosion data predict times to 10% water loss ranging from 6.3 to 364 years, with the bulk of the data predicting times in the 30 to 100 year range.

2. However, field post morta data on actual positive grid corrosion in actual VRLA cells suggest much shorter lifetimes, predominantly less than 10 years, with an overall range of 4 to 36 years.

3. Direct gas collection data for AGM cells tend to indicate much greater lifetimes than from the corrosion data. These results suggest either a flaw in our understanding of the cause/effect relationship between grid corrosion and hydrogen evolution or, more likely undetected hydrogen leakage problems.

4. The much shorter lives predicted by the weight loss measurements would seem to confirm some leakage in gas collection. For example, in the 1989 publication2', the weight loss data indicated approximately 7 years, vs. the 80 years predicted by the gas collection experiment in the same publication.

5. The Gel gas collection data all represent a single manufacturer's product and appear to be relatively consistent with our expectations as to gel aging and float voltage effects.

6. Finally, in examining the gas collection/recombination efficiency tests suggested in the various national VRLA standards, we find requirements for recombination efficiency divorced from any requirement of float current at the test float voltage. This seems to indicate a lack of appreciation of the role of float current in determining the overall rate of gas evolution at a particular value of recombination efficiency.

7. While the tests and requirements of the IEC standard are specific, the requirement allows cells to gas at rates which would lead to 4 year lifetimes.

RECOMMENDATIONS:

From the data presented, it should be obvious that a standardized method of reporting all these types of data should be seriously discussed and (hopefully) adopted by the industry. Its widespread use would significantly enhance comparison of the relative performance of VRLA designs under various conditions of use. More importantly, it should enable the industry to clarify its understanding, both qualitatively and quantitatively, of the roles of the various competing reactions which occur during operation of VRLA batteries. The standardized units presented in this paper are offered only as a suggestion, but one which can be readily understood, by all those associated with the technology, regardless of their level of expertise.

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