FACTORS AFFECTING PFC EMISSIONS FROM COMMERCIAL ALUMINUM REDUCTION CELLS

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Abstract

The US Environmental Protection Agency (EPA) and the US Aluminum Association sponsored measurements of two perfluorocarbon (PFC) gases: tetrafluoromethane and hexafluoroethane. The measurements at six primary aluminum production facilities provided data on emissions of these compounds during normal aluminum smelting operations. The measurements were made using process mass spectrometry, a technique capable of monitoring the rate of emissions with a time resolution of seconds. The PFC concentration measurements were combined with smelting process data collected during the measurements to provide new insights into the relationships between the process variables and PFC emissions. Detailed data were obtained at several locations relating overall cell voltage and PFC emission rates. The profiles of PFC emissions and voltage at a time resolution of seconds provide valuable insight into how emissions vary with voltage. The data on emission rates from commercial cells as a function of cell voltage were compared with similar data developed on bench scale experiments. Other analyses included a comparison of emission rates among different cell technologies and anode effect (AE) kill strategies. Estimated PFC emissions for different definitions of AE were compared with total measured PFC emissions. Finally, the emission rate as a function of AE duration was examined.

Introduction

Two gaseous perfluorocarbon (PFC) compounds, tetrafluoromethane (CF_4) and hexafluoroethane (C_2F_6), are emitted as byproducts of primary aluminum production. These compounds, both strong greenhouse gases, are released during the temporary condition known as anode effects (AEs) (1-3). Reduction of the frequency of AE occurrence or complete elimination of AEs is desirable considering both environmental benefits (e.g., lower greenhouse gas emissions) and economic benefits (e.g., lower power consumption, decreased manpower required to treat AEs, increased aluminum production, decreased consumption of fluoride, and longer pot life). Primary aluminum producers in the US have worked cooperatively with the US EPA to reduce emissions through the Voluntary Aluminum Industrial Partnership (VAIP)(4).

Cosponsored by the US EPA and the US Aluminum Association as one element of the VAIP, PFC measurements were made at five US and one Canadian location in 1999. The initial results of the study provided aluminum specific emission levels for both CF_4 and C_2F_6 for a range of reduction technologies including center work prebake (CWPB), Pechiney CWPB, side work prebake (SWPB), vertical stud Soderberg (VSS), and horizontal stud Soderberg (HSS) cells (5). Calculated emission slope factors in kg CF_4 or C_2F_6 per AE minute per cellday were compared with the recommended best practice values published by the Intergovernmental Panel on Climate Control (IPCC) (6). A qualitative relationship was demonstrated between emission rate and AE voltage (5).

The objective of the current work was to achieve a better understanding of the dynamics of AEs in commercial electrolysis cells. A better understanding can lead to a more efficient strategy in reducing AEs and the resulting PFC emissions. The current work provides further analysis of and conclusions from the data generated in the 1999 US EPA and US Aluminum Association measurement study. An analysis of the impact of AE voltage on PFC emission rate was made and compared with similar analyses from earlier laboratory studies (7-9). A detailed analysis of PFC emission rate as a function of time on AE was made to better understand differences in emission rates between facilities operating with similar technologies and those operating with different technology types. The effects of cell design and control strategies on PFC emissions were considered. In addition, the impact of choosing an arbitrary voltage level for defining the initiation of an AE was considered. Finally, the variation of emission rate with AE duration was examined.

Experimental

Detailed descriptions of the measurement apparatus and the associated data collection strategies have previously been published (5). In summary, concentration of CF_4 and C_2F_6 were measured by on-line mass spectrometry continuously in the collective exhaust ducts of commercial reduction cells with results averaged and reported every six seconds. Process data were collected over the period of the measurements that included AE frequency, time on AE, and, when available, voltages during AEs.

Anode Effect Mechanism

Alumina is consumed and depleted from the bath in the production of aluminum as oxygen is oxidized and reacts at the carbon anode surface to produce carbon dioxide. The concentration of oxygen containing ions in the boundary layer at the anode surface decreases rapidly as the alumina concentration decreases below 2% in the bulk bath causing the anode over-potential and cell voltage to increase correspondingly. Examples of this behavior are illustrated for major smelting technologies in Figures 1 and 2.

When the critical anode current density is exceeded, fluoride containing ions are oxidized at the carbon anode surface resulting in the formation of CF_x compounds that decompose into gaseous tetrafluoromethane (CF_4) and hexafluoroethane (C_2F_6). These gases form a nearly continuous gas film that wets the bottom anode surface causing a sharp increase in cell voltage (normally 25 to 35 volts).

Anode effects occur when the critical anode current density is exceeded. Several different cell operational conditions can contribute to this situation, including a decrease in alumina concentration, an increase in cell amperage or a decrease in anode surface area as a result of a decrease in anode immersion or frozen bath covering the bottom of anodes (e.g., start-up pots).

Side Current and Magnetohydrodynamic (MHD) Instability During Anode Effects

A higher percentage of electrical current is conducted from the sides of anodes during AEs due to the high resistance of the gas film completely wetting the bottom of anodes. The gas film at the sides of anodes is thinner and more mobile than the bottom film. As indicated in Table I, Soderberg cells have less side surface area available for current flow during AE than that available to prebake cells. Additionally, electrical current is more evenly distributed across the cell from the sides of prebake anodes during AEs.

Table I Comparison of the Difference in the Side Anode Areas Between Soderberg and Prebake Cells

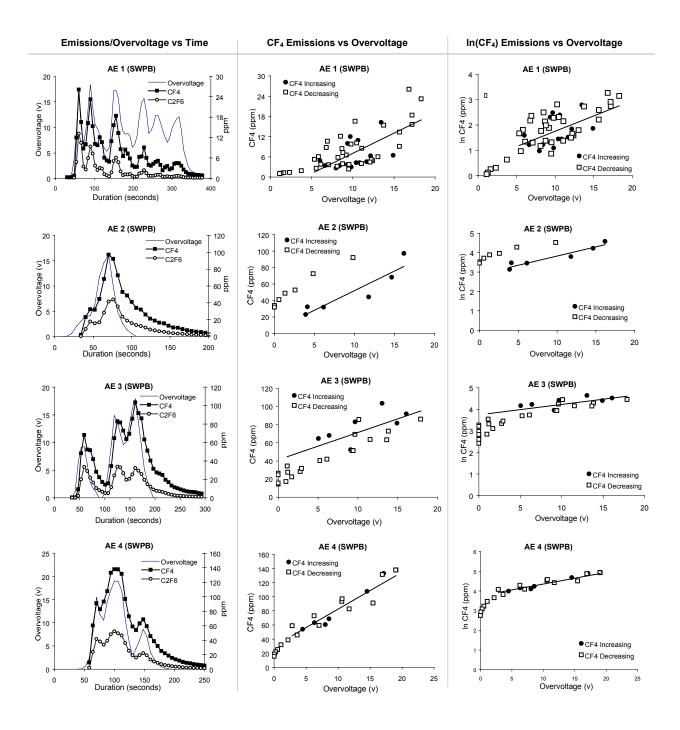| Cell	Immersion (cm)	Areas (m^2)			Side (%)
		Bottom	Side	Total	
AP-18 PB	19	23.24	14.90	38.14	39.1
Soderberg	19	23.24	5.16	28.40	18.2

Due to the limited side area, Soderberg cells generally have higher maximum AE voltage compared with prebake cells, especially when operating with lower bath levels. This can be seen in Figure 2, AE 8 , where maximum voltage during the HSS AE is higher than any of the prebake AEs.

The MHD instability increases rapidly in cells during AEs due to the large imbalance in the anode current distribution, current fluctuations in prebake anodes, and increased flow of current out of the sides of anodes.
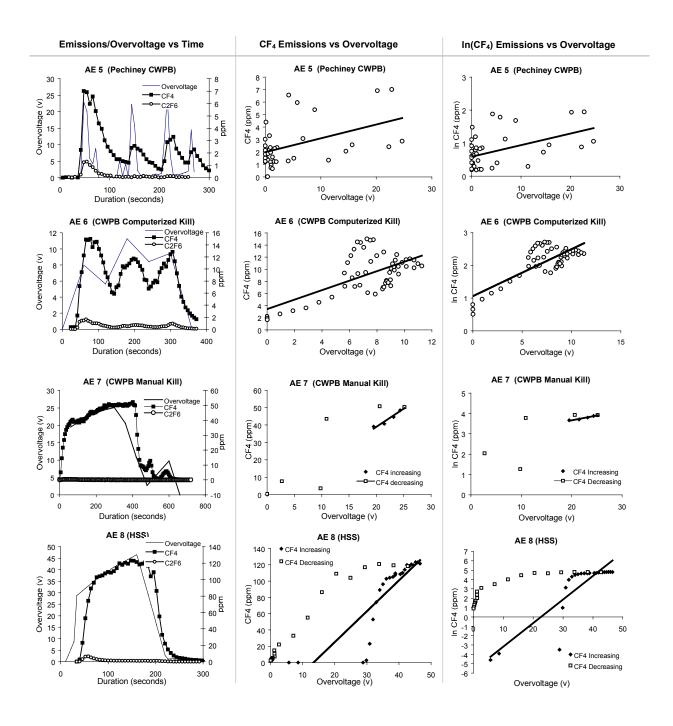
Figure 1: PFC Emission Rates and Cell Overvoltage with Time on Anode Effect

Figure 2: Comparison of Anode Effects From Different Technologies

Emissions/Overvoltage vs Time	CF$_4$ Emissions vs Overvoltage	In(CF$_4$) Emissions vs Overvoltage

Anode Effect Kill Mechanism and PFC Emissions

Anode effects are killed in cells by causing direct contact (significant electrical short) between the aluminum metal pad and carbon anodes. This is accomplished by using several methods: lowering the anode(s) until making contact with the aluminum metal; causing the metal to splash up; making contact with the anode(s) using wood poles, air lance, rakes, etc.; or decreasing the line amperage to zero.

Prebake anodes do not have to be lowered very far to make contact with the highly unstable aluminum metal pad in order to kill AEs. Also, cells that operate with either a low metal pad depth, poor magnetic designs, or large metal pad surface deformation are even more unstable due to MHD instability during AEs and are easier to kill than cells with higher metal pad depths.

Anode effects are killed very quickly (less than 30 seconds) with fast anode down-moves in prebake cells having a poor magnetic design with low or distorted metal pads and close anode-cathode distance (less MHD stable during AEs). PFC emissions stop immediately when the AE is killed. The cell voltage decreases sharply from the high AE voltage, 25 to 35 volts, and remains constant at the cell operational voltage of 4.2 to 4.5 volts as illustrated in Figures 2 and 3.

Anode effects are killed less quickly (2 to 6 minutes) with small progressive anode down-moves in prebake cells having a good magnetic design, higher metal pads and wider anode-cathode distance (more MHD stable during AEs). PFC emissions are emitted nearly continuously during AEs in prebake cells having stable metal pad conditions. During the AE, cell voltage is consistently high. When the AE is killed, the voltage decreases sharply and remains constant at the cell operational voltage. PFC emissions decrease with each anode contact (shorting) with the metal pad and increase again after the contact until the AE is eventually killed. Correspondingly, the cell voltage decreases with each contact with aluminum metal and increases after contact. The cell voltage decreases sharply and remains constant at the cell operational voltage when the AE is eventually killed.

The aluminum metal pad in Soderberg cells is highly unstable during AEs due to large MHD forces generated from the imbalance in the electrical current distribution due to the high current being conducted at the sides and ends of the anode.

The average PFC emissions per AE minute is less from HS and VS Soderberg cells compared with prebake cells due to the high degree of contact (shorting) with the metal pad. The anode cannot be lowered in Soderberg cells to kill the AE; if it were, the molten bath would be flooded out of the cell cavity. PFC emissions are emitted non-continuously in HS and VS Soderberg cells due to the highly unstable metal pad. PFC emissions stop and start with each shorting of the aluminum metal contact with the anode. Correspondingly, the cell voltage is highly unstable, changing rapidly from around 4 and 5 volts to 35 volts. Each time aluminum metal makes contact with the anode, the cell voltage decreases rapidly. When the AE is eventually killed the voltage remains constant at the normal cell operational voltage.

Voltage Effect on PFC Emissions

Measurement of PFC concentration was made in near continuous time with averages recorded every six seconds. The concentration measurements were, however, displaced in time from corresponding cell voltage measurements by approximately one minute. This delay results from the taking of the gas sample by a probe inserted in a collective duct for a number of reduction cells and the transfer of the gas in three-eighths inch diameter tubing for an additional distance of 30 to 50 meters. Consequently, a displacement in time between corresponding concentration and voltage measurements occurs. Also, the measured PFC signal is further distorted from the true emissions profile from diffusion effects in the transfer lines. A time shift was calculated by taking the difference in time between the maximum in voltage reading and the maximum in concentration measurement. This shift was then applied to all concentration time readings to put the two data sets, voltage and concentration on the same time scale.

Figure 2, AE 1, shows time trends for cell voltage and the corresponding apparent emission rates for CF_4 and C_2F_6 for an AE from a SWPB cell. The trend illustrates several points about AE behavior in smelting cells. First, the apparent emission rate, directly proportional to the instantaneous PFC concentration at any point in time, is strongly related to voltage. Second, as the duration of the AE increases, the PFC emission rate is less responsive to voltage increases. Finally, even though concentration is being recorded at six-second intervals, there is a sampling system and measurement instrument distortion in the measured PFC concentration profile. When voltage is rising, the concentration increase tends to lag behind the voltage increase. After the voltage peaks, PFC concentrations do not decrease as rapidly as the voltage decreases.
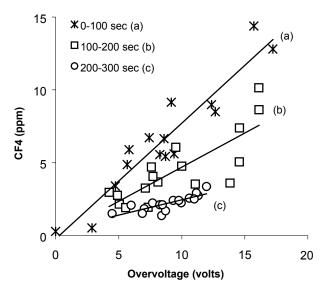

Figure 3: Apparent PFC Emission Rate vs. Cell Overvoltage

Figure 3 shows a linear increase in CF_4 emission rate with increasing cell overvoltage[1]. The emissions vs. voltage data are

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[1] Cell overvoltage is calculated as the voltage minus the base voltage where the base voltage is the interpolated voltage between the pre- and post-AE voltage.

shown in three series: (a) covering the first 100 seconds of the AE; (b) covering the period from 100 to 200 seconds; and (c) covering the period from 200 seconds to about 300 seconds. The data show that a linear relationship exists between voltage and the instantaneous CF_4 concentration over different phases of the AE 'lifecycle'. Over the 300 seconds of AE, the emission rate varies by a factor of about three from the first 100 seconds to the last 100 seconds (i.e., at the same voltage, emissions during the latter part of the AE are approximately one third of the emissions during the initial part of the AE).

After the AE is killed the voltage is somewhat lower than normal operating voltage, probably because the anodes have been lowered reducing the bath resistance. Also, the excess energy from the AE has increased bath temperature, which also reduces cell voltage. Figure 3 illustrates the relationship between voltage and emission rate for CF_4, however, the behavior for C_2F_6 follows similar trends. The small difference in potential for C_2F_6 formation, about 0.2 volts, is not evident from analysis of the voltage data, which was only available in 10-second averages from this facility.

These data show that cell voltage is the primary determinant of PFC emission rate, however, a secondary dependence of emission rate on AE duration also exists. Several possible explanations exist for this behavior. In commercial cells on AE, feeding of alumina is taking place either directly by the point feeding of alumina, or indirectly through "pumping" of anodes or "green poling" while killing the AE. This addition of alumina may then set up a competing reaction reducing the rate of PFC generation. One possible way of accounting for this factor in estimating PFC emissions is to develop facility specific factors of emission rates vs. voltage at appropriate time intervals of the AE. Modern computer control systems, which capture voltage information in high time resolution (tenths of a second), lend themselves to such active data collecting and sorting.

PFC Emissions From Laboratory and Commercial Cells

A theoretical basis has been described relating the rate of PFC emissions on cell overvoltage for laboratory scale electrolysis cells (7-9). The work predicted that PFC emission rates were an exponential function of the anode overvoltage. Bouzat et al. proposed an empirical relationship predicting CF_4 emissions as a function of overvoltage in commercial electrolysis cells (10). The latter relationship predicts a linear change in CF_4 emissions with time-integrated overvoltage. Other equations have been proposed to estimate PFC emissions based on AE minutes per cellday without taking AE voltage into account (6). Figures 1 and 2 show the relationship of PFC instantaneous concentration and cell overvoltage with time on AE. All the graphs illustrate the qualitative dependence of emission rate on voltage because peaks in PFC instantaneous concentration always accompany peaks in voltage. A time dependence is also evident. The dependence on time appears to differ from one AE to another as illustrated between AEs 4 and 5. A marked decrease occurs in the responsiveness of emissions with voltage spikes as the AE progresses in AE 5 while in AE 4 a little decrease occurs with voltage as the time on AE increases.

Figures 1 and 2 also shows for each of the AEs illustrated, the relationship of ppm CF_4 and the natural logarithm of ppm CF_4 vs.

voltage. The logarithmic relationship is expected to better fit the data than a simple linear plot if the laboratory scale work referred to previously applies to commercial cells. As can been seen in Figures 1 and 2 a reasonable linear fit exists between ln ppm CF_4 in the higher voltage ranges. Further analysis is required to determine the exact nature of the relationship (e.g., linear or exponential) between emission rate and voltage. An apparent offset exists in trendlines between data collected when PFC concentration is increasing from that when concentration is decreasing. This offset probably results from the sampling and instrument systems distorting the actual emission profile.

Voltage and PFC Emission Profiles Among Cell Technologies

The measurement data collected in the current series of measurements offer the opportunity to test the applicability of the previously mentioned relationships in a variety of commercial electrolysis cells. Analysis of the data was made at a SWPB facility, a CWPB facility, a Pechiney CWPB technology facility, and a HSS facility. Figures 1 and 2 show the strong relationship between emission rates and voltage for all technologies.

Figure 2, AE 5 illustrates the detailed effect of voltage on emission rates for a CWPB cell operating with Pechiney control. The good correspondence of times of emissions changes of both CF_4 and C_2F_6 with voltage changes is evident from the figure. As in Figure 1, AE 1 (SWPB), AE 5 indicates a decrease in responsiveness of PFC emissions after the initial voltage peak with further increases in voltage.

Voltage and PFC Emission Profiles Between AE Kill Strategies

Figure 2, AEs 6 and 7 show profiles for a CWPB cell for a computerized and manually killed AE, respectively. Both profiles illustrate the strong relationship between overvoltage and emissions. The manual kill profile is interesting in that the rate of change of voltage and emissions are relatively small after the AE is initiated. Given the steady state nature of the voltage and emissions, emissions estimated based on an average emissions factor are likely to be more successful than for computer killed AEs that are characterized by more complex voltage transitions.

Because the voltage readings for the CWPB facility were only available as one minute averages, any detailed analysis of areas where voltage and emission rates were changing rapidly was precluded. Figure 2, AE 7 also shows the relationship between emission rate and voltage. The results are similar to the previous results in that the strong linear relationship of voltage and emission rate is seen from the data between 20 and 30 volts when the voltage and emission rates are increasing.

Definition of AE Duration

Different producers have adopted a variety of conventions for defining the duration of AEs. These conventions are generally based on a "trigger voltage", i.e., the voltage above which the AE is said to have begun, and the voltage below which the AE is defined as terminated. Variations also incorporate delay times

Table II PFC Emissions From Normal Operating Voltage to 8 Volts Compared to Total Emissions for a SWPB Cell

CF_4 ppm-sec	CF_4 ppm-sec 4.3v – 8v	% of total ppm-sec

2,357	23	1
9,553	437	5
11,925	839	7
5,691	531	9
5,183	661	13
10,723	1,369	13
5,863	794	14
9,165	1,325	14
	Average:	10

after which voltage rises on the same cell are defined as a new AE rather than a continuation of the previous AE. Most producers have assumed that the time spent in the transition voltage range between normal operation voltage, around 4.3 volts, and the trigger voltage, usually between 8 to 10 volts, is not significant for estimating emissions compared with the overall duration of the AE.

Table II shows the PFC emissions for eight AEs measured for a SWPB facility. As indicated, the amount of CF_4 emissions occurring in the intermediate voltage range between normal operating voltage and 8 volts (a typical voltage defined for computer systems to start registering AE duration) ranges from 1% to 14% of total emissions, with an average of 10%. As indicated, the total time recorded by plant computer systems as AE duration based on the arbitrary "trigger voltage" may induce bias in estimating the total time during AE when emissions occur. The extent of the bias will likely vary depending on the computer control algorithm used to define the onset of the AE. Further analysis is required to quantify the magnitude of the bias in anode effect duration. This bias in anode effect duration is not expected to contribute significantly to calculations of PFC emissions either by IPCC Tier 2 or Tier 3b methodology.

Correlation of Emissions With Overvoltage and AE Minutes

Several studies indicate that integrated overvoltage is a good predictor of PFC emissions. First, Nissen and Sadoway have laid a theoretical basis for a direct relationship between PFC emissions and AE overvoltage (7). Next, the work of Bouzat showed the strong correlation of PFC emissions with integrated overvoltage (10). Finally, the analysis of individual AE profiles in this work also showed the strong correlation of PFC emission rate with voltage. However, previous analysis results of the PFC emissions data from those facilities where both voltage and AE minutes per cellday were available showed that the correlation of overvoltage was no better than the correlation of time on AE alone when both were regressed against Kg PFC/mt Al (5).

Additional analyses are needed to determine the relative predictive capabilities of overvoltage versus AE minutes per cell-day in estimating PFC emissions.

Measured and Estimated Emissions Based on IPCC Methodologies

IPCC Good Practice Guidelines document three different methods for calculating PFC emissions based on what process data is available (6). These three methods, in order from least accurate to most accurate, are referred to as Tier 1, Tier 2 or Tier 3. Two Tier 3 methods exist: Tier 3a and Tier 3b. Tier 3a methodology recognizes that continuous real time monitoring has potential for the highest level of accuracy; however, reliable, cost effective technology for carrying out full time monitoring is not yet available. Alternatively, Tier 3b methodology uses individual facility-specific emission factors based on measured emissions and AE minutes per cellday or line overvoltage. The Tier 2 method employs an average emission factor determined by averaging measured emission factors for facilities within a given technology family, CWPB, SWPB, VSS, or HSS.

Table III compares estimated PFC emissions based on the Tier 2 methodology with those based on measurements. The data in Table III show the improvement in accuracy associated with the use of facility-specific measurements, i.e. Tier 3b approach, for determining PFC emission factors rather than the use of technology-wide average emission factors. In some cases errors greater than 100% of the measured value result from use of Tier 2 methodology. Errors that result from use of Tier 2 average emission factors for calculating emissions from individual facilities will be lessened as additional measurements are made to better define the magnitude of the coefficients. Additional measurements will also better define the characteristic variability of emissions among facilities of the same technology type. Additional measurement results are needed for both VS and HS Soderberg cells as current measurements of emission coefficients show the most variability for those technologies. Also, at the time the IPCC Guidelines were written, insufficient data were available to set coefficients for C_2F_6 for HSS or SWPB technologies. A default value of ten percent of the CF_4 emissions was used for the Tier 2 coefficient. Based on measurement results documented after the IPCC Report was published more accurate Tier 2 coefficients can be derived. The use of Tier 2 coefficients to calculate emissions for individual facilities will continue to result in significant errors in reported emissions because of the inherent variability of PFC emissions among operators of similar technologies.

Table III Comparison of PFC Emissions Calculated From IPCC Tier 2 Guidelines with Measured Emissions

Facility-(Tech-Type)	Process parameter Value	Parameter	CF_4 Emissions(kg/mt Al)			C_2F_6 Emissions (kg/mt Al)		
			CF_4 meas	CF_4 IPCC - 2	% diff.	C_2F_6 meas	C_2F_6 IPCC-2	% diff.
A-(VSS)	12.6	ae min/cellday	1.5	0.86	43%	0.06	0.038	37%
B-(SWPB)	4.5	ae min/cellday	1.18	1.31	-11%	0.4	0.13	67%
C-(HSS)	2.2	ae min/cellday	0.17	0.40	-133%	0.02	0.040	-98%
D-(CWPB)	1.4	ae min/cellday	0.3	0.20	35%	0.05	0.025	50%
E-(CWPB)	2.7	ae min/cellday	0.44	0.38	14%	0.05	0.049	3%
F-(CWPB-Pechiney)	0.69	AEO (mv)	0.007	0.01	-97%	0.0005	NA	NA

Conclusion

Tetrafluoromethane and hexafluoroethane are emitted from commercial primary aluminum reduction cells when the AE overvoltage rises to levels high enough to initiate oxidation of bath fluoride components at the anode. This condition occurs primarily at AE due to low alumina concentration for commercial cells. The rate of PFC emissions is a function of the cell AE overvoltage that results from the high resistance of the inert film formed on the anode. The accurate determination of overvoltage throughout the duration of an AE is complicated by the fact that typically computer control systems are reducing anode-cathode separation in an effort to kill the AE and the cell is experiencing temperature increases due to the AE.

The rates of emissions from the data analyzed here from commercial cells fit reasonably well with prior relationships developed in laboratory cells showing an exponential dependence of emission rate with anode overvoltage. However, for some AEs, the relationship is more linear than exponential. Confounding effects resulting in distortions and scatter of the measured PFC emission peaks make it difficult to be exact about the fit of data. Consequently, additional analysis of the data is needed to determine the best fit for the data.

The strong relationships developed here between cell voltage and emission rate warrant a closer look at computer algorithms using combined voltage and time on AE to more accurately track PFC emissions from voltage process data. The apparent decrease in PFC emission rate with extended AE duration requires a computer algorithm that incorporates this factor in accounting for PFC emissions. Another relationship that deserves further study is the variability of PFC emissions per AE minute across different AEs. Understanding this variability is important because the accuracy of current methods for estimating PFC emissions is dependent on regressions based on AE minutes per cell-day. Despite the variability observed among AEs, a representative slope coefficient can be developed over a large sample of AEs for use in the IPCC Tier 3b approach.

The data analyzed here indicate that emissions occur below the trigger voltage defined for many computer systems for tabulating AE duration. The use of a trigger voltage should not affect the overall accuracy for estimating PFC emissions using the Tier 3b methodology since the bias in AE duration will be reflected in the emission factor calculated for the facility. Additional analysis is needed to evaluate the impact of a trigger voltage on estimating emissions using IPCC Tier 2 methodology, however, it is not expected to be significant compared to other sources of variability in emission factors among operators of similar technology categories.

The use of IPCC Tier 2 methodology for calculating PFC emissions from current emission coefficients can result in errors in excess of 100% of the measured value. Reports of additional measurements, particularly for Soderberg technology, will improve calculated emissions by developing more accurate coefficients and a more quantitative understanding of the variability of results from individual facilities. Measurements must be carefully made with good measurement practices including quality assurance of the measurement process. Tier 3 methodology should be used for most accurate calculation of emissions from individual facilities.

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