

ANODE EFFECT PHENOMENA DURING CONVENTIONAL AEs, LOW VOLTAGE PROPAGATING AEs & NON-PROPAGATING AEs

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Abstract

Anode effect (AE) phenomena in aluminium cells can be separated into several categories. Firstly, 'conventional' AEs (>8V) are typically initiated on one or two localized anodes and then, due to an abrupt increase in current density, rapidly propagate to the other anodes in the cell thereby providing the typical emission spectrum of PFCs. Secondly, 'low voltage propagating' AEs (<8V) result from localized AEs rapidly propagating to a limited section of anodes with the cells remaining below conventional AE voltage; these AEs often undergo electrical shorting, especially at narrow ACDs, resulting in rapid self-termination. In contrast, the continuous background emission of PFCs should be categorized as a third type of AE or 'non-propagating' AEs. The fundamental mechanisms that initiate continuous PFCs very likely still apply, but the localised AEs do not propagate sufficiently to other anodes for a cell to exhibit a voltage signature characteristic of a low voltage AE.

Introduction

Anode effects (AEs) have been a topic of extensive study and research in the primary aluminium industry due to the generation of harmful perfluorocarbon (PFC) greenhouse gases, CF₄ and C₂F₆, which have extremely high global warming potentials of 7,390 and 12,200 times equivalent of CO₂ [1]. In the past decades, the aluminium industry has been very active in targeting PFC emissions and has made considerable progress in reducing them by addressing both the frequency and duration of AEs [2].

Traditionally, PFC generation from AEs is associated with a rapid increase in cell voltage. AEs are detected by the control system when the voltage on a cell exceeds a certain threshold. For example, a commonly accepted AE definition is *when cell voltage exceeds 8V for more than 3 consecutive seconds* [3]; however, the AE trigger voltages do vary in the industry from as low as 6V to as high 10V [2]. These definitions form the basis for AE frequency and duration statistics, which are often used to estimate total PFC emissions for a plant. In this paper, the authors refer to these as 'conventional' anode effects (conventional AE).

In recent years, the discovery of PFC emissions in absence of any 'detected' or officially *declared* anode effects (AE) has sparked considerable interest in the aluminium smelting community, evidenced by numerous studies on the subject in the past years [2, 4-8]. PFCs generated in such scenarios are not declared by smelter control systems as 'anode effects' as they are either: (i) 'low voltage' anode effects that have very similar PFC emission signatures to conventional AEs, but exhibit peak voltages that fall below the threshold voltage (e.g. <8V) used by control systems, or (ii) are continuous background emissions of PFCs that do *not* appear to have the same voltage and PFC emission characteristics

as conventional (>8V) and low voltage (<8V) anode effects. While these two scenarios are sometimes referred to as 'non-anode effect' or NAE emissions [2, 4-8], there is actually mounting evidence to suggest that PFC generation in both scenarios is *initiated* by the very same fundamental mechanisms as conventional anode effects. However, the major difference is likely to be how these anode effects are *propagated* within the cell. As such, in this paper the authors define these two scenarios or categories as: (i) 'low voltage propagating' anode effects (LVP-AE) and (ii) 'non-propagating' anode effects (NP-AE).

This paper sets out to present the mechanisms behind the two alternative categories of AE. To understand these mechanisms the theory of AE initiation, propagation and termination is first reviewed.

Theory of AE Initiation, Propagation & Termination

Anode effects are typically *initiated* on a few individual anodes. Electrolysis of fluorides occurs when the current density exceeds the critical current locally, at any given location. This can be caused by a combination of: (i) insufficient levels of dissolved alumina locally and/or (ii) an increase in local current density on anodes. As anode current density increases, anode polarization voltage also increases until the anode reaches its critical current density (CCD) [9]. This results in an anode effect, as confirmed by the generation of a C-F gas film on the anode surfaces and the emission of PFC gases (CF₄, C₂F₆) [9]. During the AE initiation period, PFCs are generated despite cell voltage not exceeding the AE voltage threshold [7].

Observations of conventional AEs show that following initiation on 1 or 2 anodes, the AE is *propagated* within seconds to all anodes in the entire cell (Figure 1). As anodes go into anode effect the formation of a C-F gas film de-wets the anode thereby severely increasing the anode resistance. The electrical current is shifted to other anodes in the cell. This step-wise current increase results in an increase in current density on other anodes until they exceed CCD and go into anode effect as well, resulting in the propagation of AEs across anodes in the cell [9]. When all (or almost all) anodes are involved in the AE, the cell voltage increases rapidly to levels of 18-50V. The initiation and propagation effect is shown in Figure 1.

For conventional AEs, PFC emissions and high cell voltages continue until the AE is effectively terminated. This requires replenishment of dissolved alumina within the anode-cathode distance (ACD) and dispelling of the C-F gas films under anodes by short-circuiting anodes with the metal pad, which is done either *manually* (with a wooden pole) or *automatically* (by lowering the anode beam and reducing ACD) until metal waves contact anodes [9, 10].

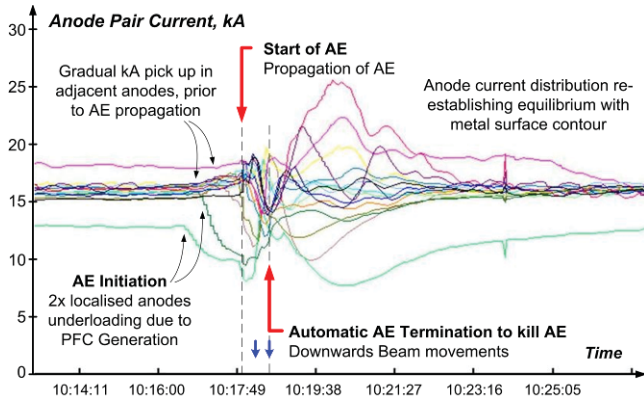


Figure 1: Individual anode currents showing initiation and propagation of a conventional AE.

Redefining Categories of Anode Effects

Three different PFC generating scenarios were presented earlier. The first is the *conventional* AE defined by control systems as when cell voltage exceeds a trigger threshold of 8V or similar. The second is the *low voltage propagating* AE that exhibits peak voltages below AE trigger thresholds. The third is the *non-propagating* AE, also referred to as the continuous background emissions of PFCs. Each category can be defined by certain voltage and emission characteristics as summarized in Table I. These are discussed in the next three sections.

Conventional AEs

Conventional AEs are those defined traditionally where the cell voltage exceeds a voltage threshold of 8V or similar. PFCs are generated as a sharp peak of emissions over time, ramping up exponentially at the start of an AE and falling rapidly back to background levels after the AE is terminated. As with PFC emissions, cell voltage rises exponentially upon AE propagation often reaching levels of 25-50V and as such they are essentially ‘high voltage propagating’ AEs. Upon termination, cell voltage rapidly drops back to normal levels (Table I) [9, 10]. Conventional AEs are generated through the process of initiation under localized anodes followed by propagation on all or almost all anodes on the cell, as illustrated in Figure 1.

Table I: Summary of the three defined categories of anode effects, showing their voltage and PFC emission signature, as well as their expected mechanisms of initiation and propagation.

AE Category	Volt Signature	PFC Signature	AE Initiation	AE Propagation
I Conventional AE	AE voltage signature > 8V	Sharp emission peak, but drops to background levels after AE termination	Initial PFC generation on 1-2 anodes due to localised anode polarisation, from a combination of low alumina concentration and high current density	Rapidly propagates to <i>entire</i> cell. Does not self-terminate most of time, requiring Automatic or Manual AET
II LVP-AE Low Voltage, Propagating	AE voltage signature 4-8V			Propagates but is <i>limited to a section of the cell</i> , e.g. 1/3 or 1/2 of cell. Self terminate after propagation.
III NP-AE Non-Propagating	No visible voltage signature	Slow moving background		Does <i>not</i> propagate, but remains on several localised anodes

Low Voltage Propagating AEs (LVP-AE)

Low voltage propagating AEs (LVP-AE) are essentially the same as conventional AEs, in terms of emission and voltage signature, however, the difference being that peak AE voltages stay below voltage thresholds. Figure 2 shows an example of the peak CF_4 and C_2F_6 emissions and corresponding cell voltage signature associated with a LVP-AE. In this example PFC emissions were measured from the duct gases of a single 400kA cell and voltage tracked at 20Hz [6]. As with conventional AEs, both the PFC emissions and the cell voltage rapidly increase and peak when the AE propagates to more anodes. However, the maximum voltage of this AE does not exceed 7V and hence is never declared an ‘AE’ by control systems. In fact, LVP-AEs with peak voltages as low as 4.2V (with normal operating voltage at 3.9-4.0V) have been noted by the authors on some high amperage cell technologies.

Figure 3 shows the distribution of individual anode currents across a one hour period that includes a LVP-AE; this is the same one hour period that is shown in Figure 2. Clearly, AE propagation was limited only to anodes towards the duct end of the pot while remaining anodes towards tap end of the pot were relatively unaffected. This *limited* propagation is seen as an explanation as to why LVP-AEs exhibit lower voltages than conventional AEs.

Another characteristic of LVP-AEs is that they typically ‘self-terminate’, in other words anodes are locally short-circuited by metal pad instability upon propagation of the AE without manual or automatic intervention. Since they never exceed the voltage threshold, LVP-AEs are never declared by control systems and hence are never terminated manually or automatically with beam movements or rapid alumina feeding.

The mechanism of LVP-AEs also points to the presence of significant alumina concentration gradients across cells that exhibit this type of AE. While areas of lean alumina concentrations are responsible for initiating and partially propagating the AE, other areas of the cell have sufficiently high concentrations to avoid exceeding critical current density, thereby limiting further propagation of the AE. The ability of LVP-AEs to self-terminate without additional feed suggests the redistribution of alumina from high to lean concentration areas in the pot, in time to prevent further PFC generation and terminating the AE.

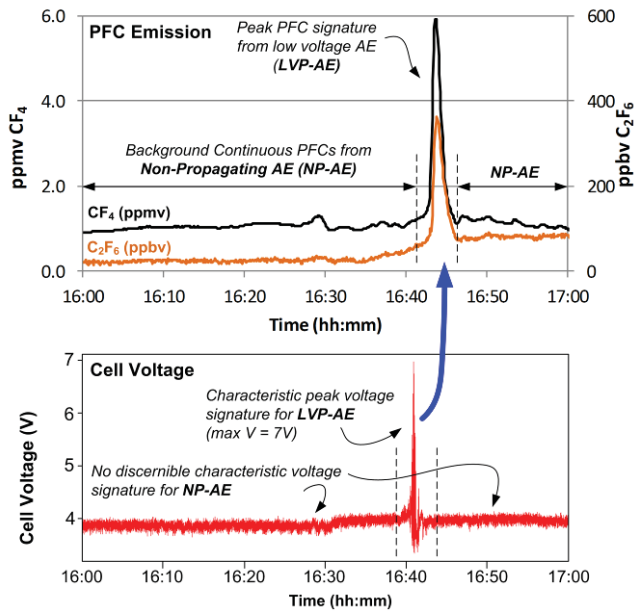


Figure 2: [Top] PFC emission and [Bottom] cell voltage signatures for LVP-AEs and NP-AEs from a single 400kA cell.

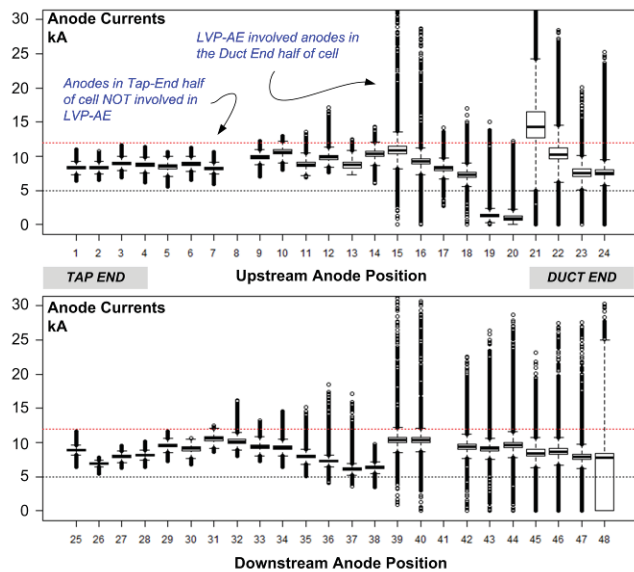


Figure 3: Boxplots of 48 individual anode currents (kA) from one cell over the period depicted in Figure 2. Note that data for anodes 8 and 41 were omitted due to faulty sensors.

'Non-Propagating' AE (NP-AE)

In this work continuous background emissions of PFCs are referred to as *non-propagating* AEs (NP-AE). These AEs have an emission signature that is very distinct from *conventional* AEs and LVP-AEs, i.e. a slow-moving background of PFCs varying in a time-scale of hours rather than short peak emissions (Figure 2) lasting only minutes. Similarly, NP-AEs do not exhibit any easily discernible voltage signatures or patterns that are normally associated with the other two AE categories. Rather, cell voltages remain at normal operating range, e.g. 3.8-4.2V (Figure 2). While

this has led to the phenomena being labelled as 'non-anode effect' or NAE emissions [2, 4-8], there is much evidence to suggest that these have the same fundamental initiation mechanisms as previous types of AEs. Several recent findings point to this.

First is the link between NP-AEs and *low alumina concentrations in a cell*. Chen *et al.* [11] showed that generation of continuous PFCs on a 300kA pot ceased when bulk alumina concentrations were above 1.8-2.0%, and being generated again when concentrations dropped below this level. During their investigations Zarouni *et al.* [7, 12] found that continuous PFCs were sometimes generated at the end of underfeed periods (about 10% of underfeeds), particularly when resistance curve slopes were higher, which points to leaner alumina concentrations in the pot. Furthermore, by adjusting the alumina feed strategies to provide a higher % alumina in the bulk of molten bath, Li *et al.* [13] were able to reduce continuous PFC background levels by 98% and 81% on 300kA and 350kA potlines respectively. This also significantly reduced the frequency of conventional AEs.

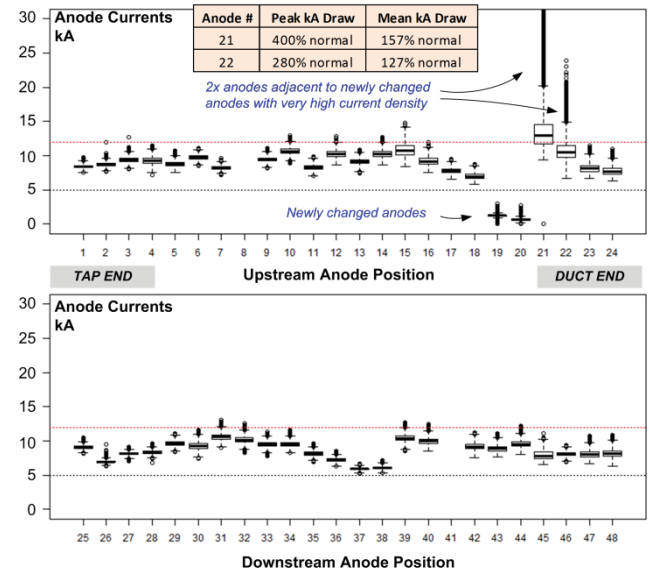


Figure 4: Boxplots of 48 individual anode currents (kA) in another one hour period with only continuous PFC emissions. Note that data for anode 8 and 41 were omitted due to faulty sensors.

Second is the link between NP-AEs and individual anodes with high current densities. Zarouni *et al.* [8] and Wong and Marks [6] both observed the generation of continuous PFCs being initiated after anode changes. These can result in colder anodes with disrupted alumina dissolution and transport, as well as an imbalance in anode current distributions, with anodes adjacent to new anodes having to take up higher current load and being more susceptible to exceeding CCD. An example is shown in Figure 4, where the two anodes (Anodes 21 and 22) adjacent to newly changed anodes exhibit massive current draws with currents fluctuating wildly and peaking periodically up to 400% and 280% normal load, respectively. Assuming a normal current density of 0.75 A/cm^2 these two anodes would be periodically drawing of up to 3.0 and 2.1 A/cm^2 , which clearly are levels that likely exceed the CCD at reduced alumina concentrations. During this one hour period continuous background PFC levels (not shown) were measured in the range of 0.5 - 0.7 ppmv CF_4 and $20 \text{ ppbv C}_2\text{F}_6$.

Furthermore, the same study found that continuous PFCs were generated in localised regions in a cell (majority of emissions from one of six feeders in one cell) rather than from an entire cell. This supports the hypothesis that continuous PFCs are only generated on a limited number of localised anodes (most likely those with high current draw). Evidence of this in limited studies by Chen *et al.* [11] also supports this.

The combination of the above studies suggest that NP-AEs are initiated by the same mechanism as conventional AEs and LVP-AEs, i.e. due to low alumina concentrations and current densities on several individual anodes exceeding CCD. However, these AEs do not propagate to other anodes in the cell, but are maintained continuously on these anodes. They are hence *non-propagating AEs*.

Factors behind LVP-AEs and NP-AEs

Having defined the three categories of AEs, the question remains as to why LVP-AEs and NP-AEs are so significant in some smelters (contributing as much as 92% of total PFCs at one plant [5]) and not in others? Are there any common factors that exist?

In this paper, it has been shown that the two types of localised AEs can occur when AEs either (i) propagate only to a limited section of the cell (LVP-AEs) or (ii) do not propagate at all (NP-AEs). There are two conditions that might limit the propagation of AEs to other anodes:

1. *Low dissolved alumina levels* under a limited section of anodes (LVP-AE) or only a few anodes (NP-AE), but higher alumina levels on all other anodes, and/or
2. *High current densities* on only a few anodes or a section of anodes in conjunction with low current densities on other anodes

Considering this, cells that have LVP-AEs and NP-AEs do appear to have a number of characteristic factors that make either of the two conditions possible. These are discussed as follows.

Large, High Amperage Cells with Low ACD

There is some evidence that LVP-AEs and NP-AEs are more prevalent for large-sized, high amperage (>300kA) cells [7]. High amperage (300-500kA) cells, such as those in China where LVP and NP-AEs were first detected, are often characterised by:

- Long cell dimensions (>18m)
- Large numbers of anodes (40-48)
- Low current densities (in the range of 0.75-0.83 A/cm²)
- Very low operating cell voltages (in the range of 3.85V) and hence low anode-cathode distances (ACD)
- Stable magnetics and hence low metal pad velocity

Note that LVP and NP-AEs are not exclusive to high amperage cells. They have also been found in 180-300kA cells in China [14], many of which target the same characteristics of low anode current densities, low operating cell voltages and low ACDs [15].

In cells that operate at lower cell voltage and narrower ACDs the local transport of alumina from point feeders to anodes is likely to be restricted. This can result in greater concentration differences of dissolved alumina around the pot, particularly for anodes the furthest away from feeders. Longer cell dimensions in high amperage cells further exacerbate the alumina transport situation.

The stable magnetics that are often characteristic of high amperage technologies also result in low metal pad velocities, which reduce interfacial stirring of liquid bath and further reduce mass transport of alumina. These conditions all increase the risk of large gradients in alumina concentrations (high concentration vs. lean zones around a pot) thereby making these cells more susceptible to localised PFC generation.

In comparison, lower amperage cells typically have better alumina transport factors (smaller cell dimensions, larger ACD, higher metal pad velocity) resulting in more even alumina distributions. As such, when AEs initiate on these cells, they typically propagate across the entire cell resulting in a conventional AE.

Cells with low ACDs also have the situation where anodes are closer in proximity to the metal pad. This makes it easier for metal pad waves to come into contact with anode surfaces, particularly when the pot is unstable during an AE. This also helps explain the tendency for LVP-AEs to ‘self-terminate’ in these cells.

Effect of More Anodes in Large Cells

The large number of anodes in high amperage cells also increases the susceptibility of a cell to localised PFC generation. When combined with low nominal current densities, a high number of anodes reduce the impact on cell voltage and average current density when AEs occur on only a few individual anodes. To illustrate this, a simple electrical balance was used to model the effect of average current density (Figure 5) and cell voltage (Figure 6) from increasing numbers of anodes on AE. Individual anodes were considered as independent parallel resistances in a cell. This was done for 200kA (with 20 anodes) and 400kA (with 40 anodes) cell scenarios, assuming a nominal current density of 0.75 A/cm². Please note that these are only simplistic models for the purposes of demonstration and numbers from the model should be used with care.

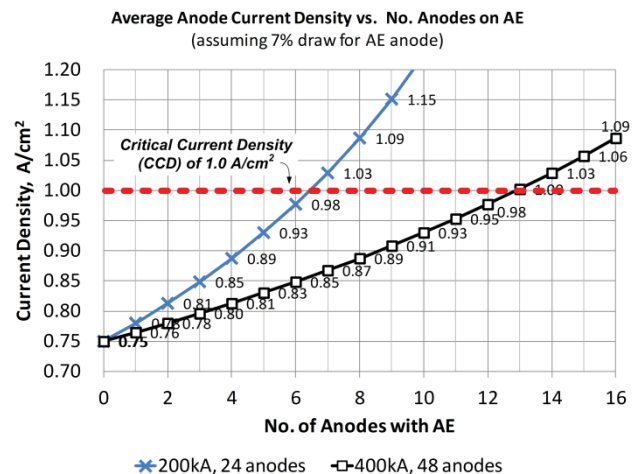


Figure 5: Model of current density vs. number of anodes on AE.

The effect on current density is shown in Figure 5 and assumes that an anode on AE draws only 7% of its normal current (due to insulating PFC gas films covering anode surfaces). In reality, the current draw of an anode on AE is dependent on a number of factors including bath levels, PFC film coverage, surface contact area of bath to anode, and so forth. An arbitrary critical current density (CCD) of 1.0A/cm² is also applied.

Figure 5 shows that on a 200kA pot, the CCD is exceeded by remaining anodes on the pot when more than 6 anodes are on AE, resulting in propagation to the entire cell. However for a 400kA pot, 13 or more anodes on AE are required for the same effect to occur. This simple example illustrates very clearly that it is *more difficult* for a large cell with high numbers of anodes to have a conventional AE. Conversely, the likelihood of an AE remaining limited in propagation, resulting in a LVP-AE or NP-AE, *increases* when a pot has more anodes. Note that in practice, anodes adjacent to those with AEs will exhibit higher current densities than the ‘average current densities’ modelled in Figure 5. Nevertheless, the general concept holds up well.

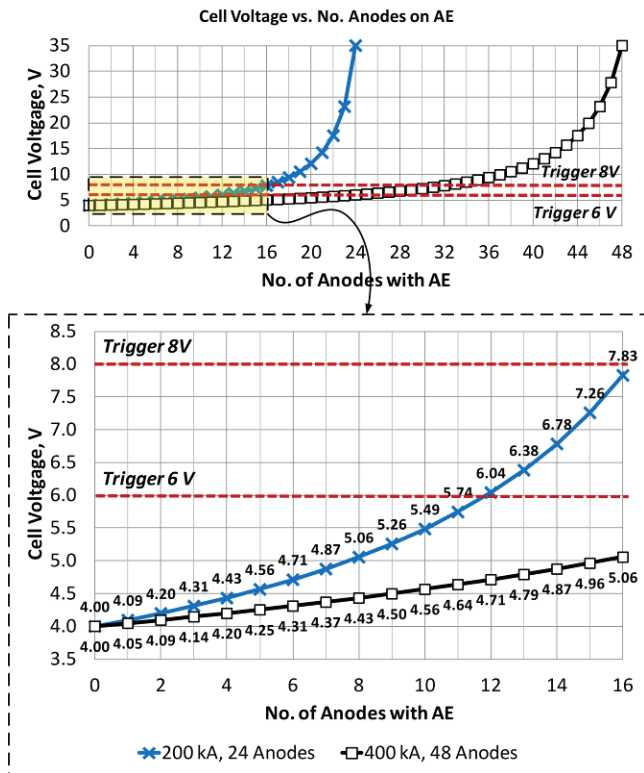


Figure 6: [Top] Cell voltage as a function of no. anodes on AE and [Bottom] a zoom-in on this.

Figure 6 shows a much greater change in voltage for a 200kA cell than for a 400kA cell for the same number of anodes on AE. The smaller effect on voltage explains why for a large cell with many anodes, continuous PFC generation can occur on 1-2 individual anodes without any perceptible change in overall cell voltage (Figure 2, bottom). The same observation holds for conventional AEs just prior to propagation. In a case study of ‘near AE events’, Zarouni *et al.* [7] showed how PFC initiation and underloading of current draw on 2 anodes resulted in a cell voltage increase in the range of only 100mV.

If an AE propagates to a limited section of anodes – for instance 12 anodes – voltage on a 200kA pot would increase to 6.04V, while on a 400kA pot, voltage would only increase to 4.71V. While 12 anodes on AE might tip a 200kA pot over the CCD limit for all remaining anodes (Figure 5), a 400kA pot might still be able to sustain 12 anodes on AE without propagation to the rest of the cell. This illustrates the principle of how LVP-AEs with

significantly lower peak AE voltages can occur on larger pots and similarly, why they are less common for smaller pots.

PFC Emissions for LVP-AEs and NP-AEs

Emissions of CF₄ per cell/day can be predicted by the equation,

$$\text{kg CF}_4/\text{t Al} = \text{slope factor} \times \text{AE minute/cell.day} \quad (1)$$

where the *slope factor* is the rate of emissions during an AE assuming that all anodes are on AE [16]. However, if an AE is not propagated to all the anodes in the cell then the rate of CF₄ emissions is likely to be proportional to the number of localized anodes on AE. A simple model exploring this concept is shown in Figure 7 for a 400kA cell with 48 anodes. This uses the typical slope factor of 0.14 kg CF₄/t Al per AE min/cell day for point feed prebake cells (IPCC Tier 2 slope coefficient [16]). Assuming an AE frequency (AEF) of 0.35 AEs/cell.day and a duration (AED) of 0.5 min/AE then PFCs would be emitted at a rate of 0.025 kg CF₄/t Al when all anodes are on AE (conventional AE).

However, if an AE is only propagated to a limited section of 20 anodes (LVP-AE) then the slope factor would drop to only 0.058 kg CF₄/t Al per AE min/cell day. Assuming the same AEF and AED parameters, PFCs would be emitted at a rate of 0.010 kg CF₄/t Al (Figure 7). This suggests that localised LVP-AEs should emit *less* PFCs than conventional AEs for the same AE frequency and duration and be proportional to the number of anodes on AE.

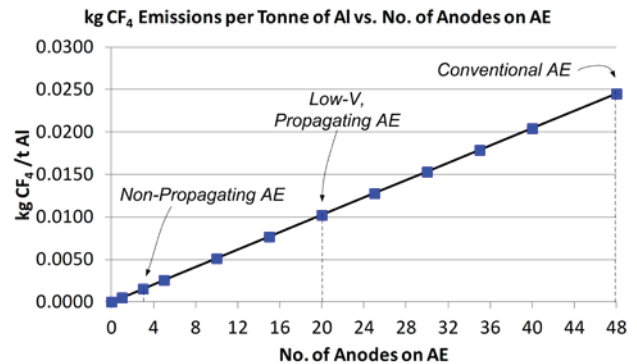


Figure 7: Simple model of CF₄ emissions per cell as a function of no. of anodes on an AE for a 400kA cell with 48 anodes.

Similarly, if an AE remains localised on 3 anodes and does not propagate (NP-AE) then the slope factor would further reduce to 0.009 kg CF₄/t Al per AE min/cell day. Assuming the same 0.35 AEs/cell.day frequency as conventional AEs it would take only 8 minutes of continuous PFC emission for the 3 anodes to generate an equivalent amount of CF₄ as if all anodes in a cell were on AE. Since these AEs often do not terminate quickly but vary over a time scale of hours, this calculation illustrates the potential for non-propagating AEs to be major contributors of a smelter’s total PFC emissions.

Terminating & Preventing Each AE Category

Terminating conventional AEs is well understood within the industry. The well-defined voltage signature is easily detected by most control systems and improvements in the automatic

termination sequences over the years have led to significant reductions in AE durations (well under 30 seconds possible [7]).

Low voltage propagating AEs can still be detected by voltage signatures but requires adaptation of the trigger. Dubal's 'near-AE' event trigger is one example, using a combination of slope and lower voltage triggers [7]. While these AEs are often observed to be self-quenching (especially in low ACD cells), they are likely to re-occur unless the cause is removed.

Conversely, non-propagating AEs are difficult to detect using only the cell voltage signal. Continuous individual anode current measurements would likely be the best method to trigger these events, while giving the location of the problematic anodes at the same time. Termination and extra feeding could then be targeted to particular problem areas.

More importantly, reducing all three AE types would be more effectively achieved by preventing the initiation mechanism from occurring. This requires maintaining all anode current densities below the CCD by control of current distribution and alumina concentration under each anode. Some key strategies include:

- Better feed control, ensuring sufficient alumina is replenished under all anodes throughout the feed cycle.
- Better maintenance of breakers and feeders, ensuring that each alumina dose gets down to the electrolyte.
- Adjusting feeding according to alumina transport around the cell.
- Ensuring that operations (e.g. anode change) do not force anodes to exceed their CCD.
- Ensuring that dissolution conditions are adequate throughout the cell by having sufficient superheat, especially after anode change.
- Aiming for bath chemistries with less modifiers for better alumina solubility. This is an increasing challenge in highly modified electrolytes [17].

Conclusions

In addition to conventional AEs, two new categories of AEs have been defined, namely: low voltage, propagating AEs (LVP-AE) and non-propagating AEs (NP-AE). The former relates to AEs that propagate only to limited sections of anodes in a cell, whereas the latter relates to AEs that do not propagate but remain localised on several individual anodes, thereby generating continuous background levels of PFCs. Cells that generate these AEs tend to be high amperage cells with large cell dimensions, squeezed ACD, low current densities, stable magnetics and a large numbers of anodes. These factors contribute to greater alumina concentration gradients and a greater propensity for AEs to remain localised. PFC emissions during LVP-AEs are lower than for conventional AEs for the same AE duration and frequency. In contrast, PFCs from NP-AEs have the potential to become the major components of total PFCs as they have significantly longer emission time-scales than the other two AEs. Strategies to terminate or reduce both LVP and NP-AEs were also proposed.

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