

SMELTER GRADE ALUMINA QUALITY IN 40+ YEAR PERSPECTIVE: WHERE TO FROM HERE?

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Abstract

All commercial Smelter Grade Alumina (SGA) is produced by Calcination of Aluminium Hydroxide in Alumina Refineries. The Aluminium Hydroxide, or Hydrate, is precipitated from a super saturated solution and classified into fine seed and coarse product for Calcination in rotary kilns or stationary calciners.

Over the past 40+ years significant technology changes/shifts has taken place in Refineries and Aluminium Smelters, the only customer for SGA. The drivers for these changes were and still are Economy of Scale, Energy Efficiency and/or improved Environmental performance.

Following an introductory review of the major technology changes/shifts that has taken place, the associated challenges in SGA quality is reviewed. Emphasis will be on Alumina properties from Calcination in Rotary Kilns and Gas Suspension Calciners (GSC) at low or high temperature onwards into Smelting, such as: Particle Size, Strength and Dust Generation & Management, HF Generation and Capture, Alumina Feeding and Dissolution. Finally the impact of major chemical impurities will be discussed. The current SGA quality requirements beyond consistency are summarized with a view to optimization of the available Hydrate Precipitation and Calcination technology. Future SGA quality requirements are discussed from a Calcination point of view as the intermediate processing step between the Refiner and the Smelter, including its potential constraints or drawbacks.

1. Introduction to Major Technology Changes

1.1 Alumina Refinery / Bayer Process

The major technology changes to the Bayer process are summarized in Table 1 below.

The increase in refinery capacity has been and is driven by economy of scale outside China. In China replication of approximately 500,000 tpy standardized trains is the preferred business model [1].

The reduction in refinery energy consumption is mainly caused by indirect slurry heating by steam in digestion, introduction of co-generation power plants and introduction of stationary calciners replacing rotary kilns.

In digestion the tube digestion technology has been introduced treating bauxite with a mixture of gibbsite and boehmite, or diasporic type bauxite [2].

The introduction of flocculants, feed slurry dilution and optimized feed well design has resulted in the introduction of much smaller tank sizes for Deep Cone Thickeners and Pressure Decanter by Rio Tinto Alcan and High Rate Thickeners and Washers by FLSmidth [3].

The Red Mud Pond incident in Hungary in October 2010 has highlighted the need for a change in design of red mud disposal areas from Dry Stacking introduced by British

Aluminium [4] to more Dry options named Dry Storage. This design change involves the replacement of Vacuum Drum Filters with Automatic Pressure Filters and transportation to the pond area with modern material handling equipment [5].

Parameter/Year	~ 1970	~ 2010
Refinery Capacity	600 -800 ktpy	1.6 - 2.4 Mtpy
Energy Consumption GJ/ton	Bayer Refinery: 13 - 14	Bayer Refinery: 8 - 9
Digestion	Autoclaves	Tube Digestion
Precipitation	Batch / Continous	Continous
Classification	Thickeners	Hydro-clones
Settlers, Flocculants Footprint Effluent	No 1.8 - 2.3m ² /tpd 100-200 ppm O/F	Yes 0.5 m ² /tpd < 50 ppm O/F
Washers, Flocculants Footprint Effluent	No 0.9 - 1.1 m ² /tpd 300-400 gpl U/F	Yes 0.25 m ² /tpd > 500 gpl U/F
Red Mud Residue	Dry Stacking British Alum.	Dry Stacking/ Storage
Calcination	Rotary Kilns & Fluid Flash (FF)	Stationary calciners FF, CFB, GSC
Particulate Emissions mg/Nm ³ (dry)	Electrostatic Precipitators 50	Fabric Filters / Bag House 20
SGA Quality	Floury & Sandy	Sandy with Low Alpha

Table 1: Technology Shifts - Bayer Refinery

1.2 Calcination Technology

In 1935 the first rotary kiln for calcination of Aluminium Hydroxide with a capacity of 110 tpd alumina was installed. In 1972 the largest Ø 4.3 x 122 m rotary kiln was contracted with a capacity of 1400 tpd sandy alumina, this technology dominated the industry.

However, in 1952, Alcoa commissioned their first 300 tpd Fluid-bed calciner producing sandy alumina [6]. Since the oil crisis in 1972 only stationary calciners have been installed (Figure 1) owing to about 25% less fuel consumption per ton alumina compared to Rotary Kilns.



Figure 1: 3 x 4500TPD GSC Units & Bag Filters

Almost all rotary kilns has now been replaced with stationary calciners and many of the latest Gas Suspension Calciner (GSC) units are now equipped with Bag House/Fabric Filters (Figure 1) instead of Electrostatic Precipitators (ESP). The major reason being that ESP units are not an absolute filter, because a plume of alumina dust is emitted in case of a power failure. In addition hereto Bag House/Fabric Filters offer a lower dust emission at lower Capex.

Around 1970 Smelters were required to lower their emission of HF by installing Dry Scrubbers requiring sandy alumina wit SSA of 50 – 80 m²/g.

Rotary Kiln Alumina	Floury	Sandy
SSA (m ² /g)	5	50-80
Alpha Alumina (%)	80-90	10-25
% Alumina < 45 μm	45-60	Max. 10
Angle of Repose	45°	30-32°
Bulk Density, Loose (kg/L)	0.8	0.95
Precipitation Yield, g Al ₂ O ₃ /L	70	50 - 80+

Table 2: SGA from Rotary Kilns

The change in alumina quality to Sandy made Floury alumina (Table 2) unacceptable to most Smelters overnight. This became a significant challenge for the Bayer Process operator in Europe to change their precipitation circuit to produce a coarser Sandy alumina particle, while maintaining the high precipitation yield of producing Floury alumina [7]. Today most modern precipitation circuit designs comprises an agglomeration section followed by a growth sections followed by a three-step classification of hydrate into coarse product for calcination, coarse seed subject to growth and fine seed subject to agglomeration. The change to sandy alumina increased the rotary kiln capacity with about 25% and lowered the specific heat consumption from about 4,315 KJ/kg floury alumina to 3,935 KJ/kg sandy alumina.

1.3 Smelter Technology

The basic smelting process is taking place in the reduction cells of which a few basic designs has evolved over time. Point Feeding Pre-Baked (PFPB) anode cell has gained a dominating market position compared to the Vertical Stub Soderberg (VSS) cell, the Centre Worked Pre-Baked (CWPB) anode cell, the Horizontal Stub Soderberg (HSS) cell and the Side Worked Pre-Baked (SWPB) anode cell.

The development of the Key Performance Indicators of the reduction cell performance is summarized in below (Table 3).

Parameter	1945	1995	2009-10
Cell Amperage, kA	25 - 50	300	200 - 500
Cell Voltage, Volt	5.0	4.1	3.85 - 4.20
Current Efficiency, %	80 - 85	92 - 95	91 - 96
Power Consumption, kWh/kgAl	20 - 25	13	12.5 - 13.5
Kg/ton Al Anode Cons.	< 776	420-550	<400

Table 3: Technology Shift in Reduction Cells

The change in basic cell design to PFPB cells and their size has been the major smelter technology change together with its associated modification of anode and cathode designs.

The pot/cell size and production have increased as indicated by the increase in cell current, while cell voltage, current efficiency and power consumption has shown only minor improvements since 1995.

With respect to the environmental performance significant improvements have taking place with respect to reduced anode consumption, number of anode effect per cell/pot per day which is < 0.08-0.1 #/p/d, total fluoride emission which is < 0.35-1.0 Kg/ton Al and average PCF gas emissions which is reduced to ≤ 0.6 ton CO₂ equivalent/ton Al [8].

2. Smelter Grade Alumina Challenges

The understanding and adequacy of the below SGA parameters (Table 4) are still under debate [10, 11].

Property	Importance [9]
Purity	Na and Ca for AlF ₃ consumption, other impurities.
Loss on Ignition	0 – 300°C adsorbed readily releases volatiles (can form “vulcanos” for point feeders). 300-1000°C predominantly Al-O-H compounds that are major contributors to HF evolution.
Bulk Density	Accuracy of correlation between volumetric feed and assured mass.
Angle of Repose	Covering quality if using only Al ₂ O ₃ , flow properties, ability to fill storage vessels actual volumetric feed transfer
Particle Size Distribution (PSD)	Consistency of properties, tendency to release dust (fines), cell performance perception of impact on solubility (coarse).
Surface Area, BET	Potential capacity for HF gas absorption.
Alpha Content	Measure of conversion of the calcined Al(OH) ₃ to the most stable alumina phase, secondary measure of surface area for a given calcination method, crusting tendency.
Attrition Index	Tendency for the poly-granular alumina to disintegrate to a finer PSD.
Flow Funnel Test	The flow ability of alumina.
Dust Index	Qualitative assessment of ability to become suspended in the cell environment and be transported away.

Table 4: Smelter Grade Alumina (SGA) [9]

New and more sophisticated parameters maybe introduced in the near future. It is however important to bear in mind that variation in SGA properties from the refinery is changed, when the SGA is handled and transported, and passes dry scrubbers [12,13,14] at the Smelter. In addition hereto improper pot operation and alumina feeding may cause more variation than variation in SGA properties from the refinery [15]. All these factors make it difficult to correctly interpret changes in pot operation from changes in alumina properties [16] as the SGA cannot speak.

The following discussion will focus on the most important aspects of SGA quality parameters and their perceived impact on smelter cell operation.

2.1 Particle Size, Strength and Dust Generation & Management

Increase in Alumina Attrition Index may lead to increased Particle Breakdown in the Smelter and thus increased content of Fines which reduces the flow ability of alumina, subject to he change in PSD that reduces the point feeding rate [17].

Alumina particle size, strength and breakdown determine the ultimate PSD fed to the reduction cell and thus its dust generation potential [17].

FLSmidth R&D work [18] has shown that larger particles break more than smaller particles when exposed to the same carrying gas velocity.

This difference is understandable as the largest particles has the highest impact energy, but the grains making up the particles exhibit almost the same chemical bonding strength regardless of size.

The particle breakdown itself has been found to depend on the parameters AAI and U [18]:

$$(1) \text{ Particle Breakdown (45 } \mu\text{m)} = f(\text{AAI} * U^2)$$

Where AAI is the Alumina Attrition Index (%) at 45 micron sieve (Alcoa method) and U (m/sec) is the gas velocity into the test cyclone.

The AAI itself depends on the morphology of the hydrate particles produced in the precipitation circuit in the alumina refinery [18].

The hydrate precipitation parameters important for obtaining a strong alumina particle with low AAI (say < 10% on 45micron) has been thoroughly studied [19]. Also the importance of avoiding over coarsening of the hydrate leading to increased particle breakdown rather than increase of the size fraction above 45 micron is of importance.

In conclusion it can be said that it make very little sense to produce a coarse but weak alumina particle (say, AAI > 20%) that breaks down relatively easy on its way to the reduction cells at the Smelter.



Figure 2: Gas Suspension Calciner

In the alumina refinery dust is mainly generated in Calciners and collected in ESP's or Fabric Filters (Figure 2).

GSC Alumina	Gibb-Site wt%	LOI (*) Wt%	Alpha %	SSA ² m ² /g
Dust	13.0	1.15	56	34.7
SGA	0	0.62	3	80.6

Table 5: GSC Dust versus SGA incl. GSC Dust

Degree of Calcination of Dust from ESP or Bag House deviates significantly from the SGA produced in the Calciners (Table 5). Smelters want to avoid dust and consequently it is of importance to consider what Dust Management Options is available in the Alumina Refinery?

- Recycle Dust to Precipitation [12] or Digestion losing Refinery Productivity and increasing OPEX.
- Mix Dust with alumina from the Fluid-Bed Cooler producing SGA with Gibbsite content?
- Recover coarse dust by Dynamic Separation and Recycle mainly Superfines to precipitation or Digestion,

whenever Precipitation experiences a Fines Generation Cycle?

- Back – Mixing Dust into the Hot Back-end of the Calciner, calcining the Gibbsite and thus eliminate it from the SGA.

The last option is successfully practiced by FLSmidth producing SGA with no measurable Gibbsite (Figure 3) and low Alpha alumina phase (Figure 4) together with significant amounts of amorphous phase:

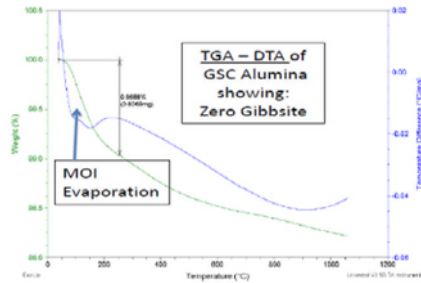


Figure 3: SGA from GSC with 0% Gibbsite

Some workers [20] is of the opinion that amorphous alumina phase is an advantage over crystalline phases with respect to the dissolution rate of alumina in the electrolyte bath.

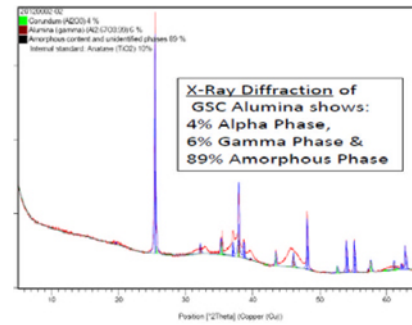


Figure 4: SGA from GSC low Alpha Phase

2.2 Generation and Capture of HF

The Specific Surface Area (SSA) is the primary physical property of the alumina specified for capturing HF gas in the Dry Scrubbing / Gas Treatment Centres emitted from the smelting pots.

The SSA increases sharply with the calcination temperature up to about 400°C [18], where after the SSA decreases to the 70-80 m²/g out of the calciners. At the same time LOI (300-1000°C) decreases from about 34.6% to below 1.0%.

It has been shown [21] that Residual Hydroxyl groups account for more than 50-70% of the continuous water released during the smelting process. Gibbsite, physically adsorbed moisture and some chemisorbed water molecules is released below 300°C [22] during heat-up of the alumina fed to the reduction cell.

In the Dry Scrubber / Gas Treatment Centre HF is chemisorbed on the SSA of the alumina [23] as per the below reaction scheme (Figure 5).

The rate of chemisorption of HF on the SSA of the SGA may be rate limited by pore diffusion subject to the duration of the contact time with SGA in the modern Dry Scrubber / Gas Treatment Centre.

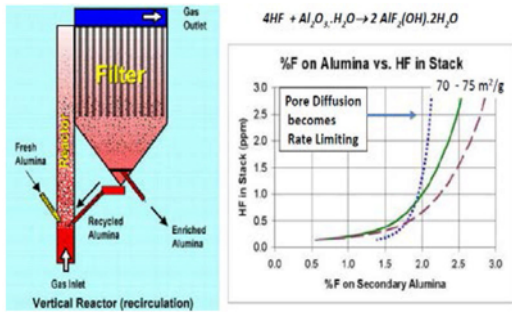


Figure 5: HF chemisorption on SGA [23].

At low gas density or quite small pores, Knudsen diffusion [24] occurs and for a strict round pore, r_e [cm] the Knudsen diffusion coefficient is:

$$(2) D_K [\text{cm}^2/\text{sec}] = 9700 r_e (T/M)^{0.5}$$

The development of pore size during Gas Suspension Calcination (GSC) is shown in below (Figure 6):

The pore size distribution of alumina from GSC units is mono disperse with an average pore size of 6 – 9 nm. The range 6 - 8 nm is believed to be optimal [16], though still not proven to be of importance [10, 16] for the HF adsorption capacity of alumina. Here it is worthwhile to remember that the molecular diameter of HF and H₂O molecules respectively, is of the order of 0.1 nm [25] and 0.2 - 0.3 nm, [26] only.

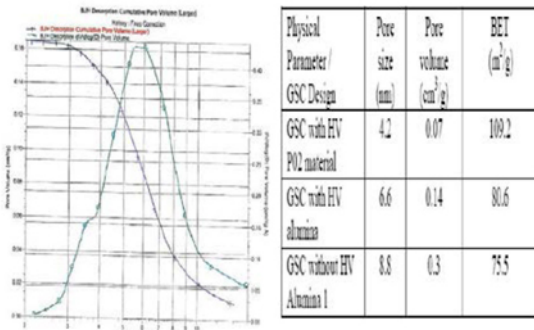


Figure 6: Pore Size Development in GSC.

HF adsorption on plant samples is correlated with SSA, though not totally linear like for laboratory calcined samples from the same hydrate source [27].

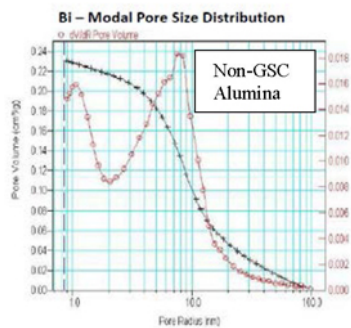


Figure 7: Calciner with Hydrate by-pass [10].

Anyway, maximizing SSA adsorption capacity can be achieved by producing alumina with a mono dispersed pore

size distribution (Figure 6), by avoiding by-pass of the Calcination furnace of hydrate, or pre-heated alumina.

The reason for the bi-modal pore size distribution (Figure 7) is due to mixing of an under-calcined alumina fraction with small pores by-passing the Calciner furnace (Figure 8), with an over-calcined fraction of alumina coming from the Calciner furnace with a large pore size but relatively smaller SSA.

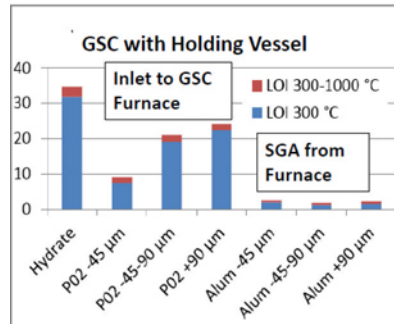


Figure 8: LOI before and after GSC Furnace.

Another way to increase the SSA sorption capacity is by lowering the calcination temperature [35].

The drawback or limitation of this modus operandi is that LOI increase with SSA and thus even more HF is generated during dissolution of the alumina requiring even more SSA. In other words Mother Nature sets her boundaries.

2.3 Alumina Feeding, Dissolution and Crusting

Impact of Cell conditions on alumina dissolution is shown below [9].

Cell Conditions	Impact on Dissolution of Alumina [9]
Mixing / Electrolyte Flow	More than a twofold impact on dissolution rate is observed by having good agitation that avoids aggregation – Poor mixing gives poor heat transfer and the combined effect gives extremely slow dissolution.
Alumina Concentration	With the normal operating range, increasing the alumina concentration can give 50% decrease in dissolution rate. However, at very low alumina concentrations, this impact is negligible – This is consistent with the basic dissolution equation and localized interfacial conditions.
AlF ₃ Concentration	In the normal operating window of reduction cells (6-14%), this has been found to have 50% impact on the dissolution rate – The combination of low operating temperature and high AlF ₃ concentration is worst.
Heat Transfer	Of similar importance to electrolyte agitation, poor heat transfer increases the tendency to form frozen aggregates. Worse for larger additions – High superheat helps reduce the extent of freezing.
Feeder Design	For fast dissolution it is important to have a steady, dispersing flow rather than “rapid” dump which can clump. – Keeping feeder holes open aids dissolution.
Mass of Feed	For a given accessible electrolyte volume, the smaller the the total mass of addition the faster the dissolution – Linked with both mixing and heat transfer.

Table 6: Impact of Cell Conditions on Alumina Dissolution [9].

Alumina dissolution rate in the liquid melt and Crust formation in the reduction cell is of primary importance for an economic (high current efficiency) and environmentally friendly (low frequency of anode effects) smelter operation [28].

Poor dissolution of alumina/Crust formation has many causes. Beyond the SGA quality coming out of the refinery (Table 4) both alumina handling and feeding system and Dry Scrubber/Gas Treatment Centre may play a significant role [14] in changing the PSD of alumina arriving at the reduction cell.

Single alumina particles dissolve in seconds regardless of degree of calcination i.e. Sandy or Floury, or Gamma or Alpha phase [29].

Alumina feeding method and rate or quantity per unit time depends on the design of the reduction cell [17] and determines the dissolution time to a large degree. Below (Figure 9) shows the effect on bath temperature for PFPB reduction cell and a CWPB reduction cell [30].

Consequently the design, operation and control of the feeding system are of paramount importance to obtain good dispersion of alumina to minimize the temperature drop and thus superheat requirements.

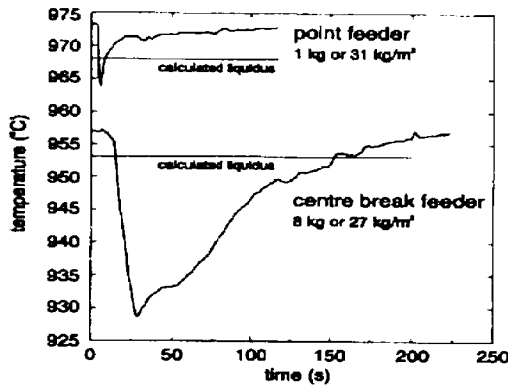


Figure 9: Temperature drop of Liquid Bath [30].

Alumina from Stationary Calciners with alpha phase content, say below 5-10% will require less superheat of the liquid melt than Sandy alumina from a rotary kiln with say 15-25% alpha content [31] at the same LOI.

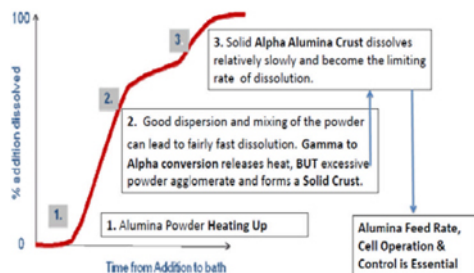


Figure 10: Dissolution Profile of Alumina [17].

Dissolution follows three distinct phases [17] as shown below (Figure 10):

- i. Heating up by cooling the bath;

- ii. Fast dissolution if good dispersion has been achieved.
- iii. Slow dissolution of alpha crust formed owing to excessive feeding or poor dispersion.

Before heat and mass transfer between alumina particle(s) and melted electrolyte can take place efficiently, the alumina particle needs to be dispersed in and wetted by the electrolyte bath. The dispersion is assisted by a relatively high LOI.

However, the amount of electrolyte bath to wet the whole particle surface per unit volume of alumina particle(s), a_v , increases with decreasing particle size:

$$(3) \quad a_v = 6 / d_p \text{ (m}^2/\text{m}^3\text{)}$$

It is therefore understandable that ESP/Bag House dust with $a_v = 883 \text{ (m}^2/\text{dm}^3\text{)}$ requires much more liquid bath per unit volume or weight than SGA including dust with $a_v = 78 \text{ (m}^2/\text{dm}^3\text{)}$ to be wetted assuming perfect dispersion into the bath. The question then becomes, why not control alumina feed rate in proportion to a_v rather than mass (bulk density), as a_v is calculated by the laser analyser used for determining the PSD anyway [32]?

2.4 Impact of major chemical impurities

Sodium from the SGA represents the largest chemical impurity introduced into the reduction cell and the bath generation rate increases with the cell size [33].

The Na_2O content in SGA from various refineries averages about 0.40% and it is more expensive to produce excess Na_2O than it can be sold for [33]. What can the refiner do to mitigate this problem?

Produce a Hydrate Particle that, when calcined has alumina attrition index less than 10-15% and about 0.30 wt% soda [34], which seems to be the equilibrium value in modern pots [20]?

Lower Soda content in the SGA, can be obtained from lower super saturation, but will reduce Refinery Yield & Productivity.

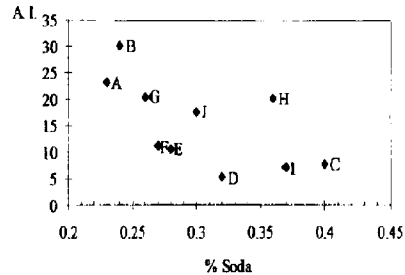


Figure 11: Alumina AI versus %Soda in SGA [34].

In addition hereto, decreasing the Soda content will also increase alumina Attrition Index (Figure 11) making a weaker alumina particle with a higher potential for particle breakdown on its route to the reduction cell [34]. So once more Mother Nature defines her limits.

However, Soda becomes Leachable after calcination and can be partly removed by a Leaching & Drying Process [33].

Can or will the Smelter justify the higher alumina price to cover the additional Refinery cost for reducing the soda level in SGA?

3. Conclusion

In view of the above the following conclusions and path forward from here is suggested:

- i. Refineries shall produce a smaller hydrate particle which upon calcination becomes strong with a low Alumina Attrition Index, say < 10-15% by Alcoa method.
- ii. Calciners shall be designed and operated with dust recycled to the hot part of the cooler system for calcining Gibbsite in the dust and minimizing hydrate by-pass.
- iii. Calciners shall be designed and operated to produce alumina with a mono disperse pore size distribution by avoiding by-pass of the Calcination furnace.
- iv. SSA/LOI relationship in Calciners shall be optimized to balance desirable SSA without excessive HF formation from too high LOI.
- v. Reduction cell feed control shall be based on the specific particle surface area $m_v (m^2/m^3)$ rather than Bulk Density to match alumina feeding with wetting capacity of the liquid bath.
- vi. Smelters consider paying a premium for SGA with equilibrium soda content and low attrition index to avoid excessive bath production and minimizing particle breakdown between Refiner and Smelting pots/cells.

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