Thermodynamics of Electrochemical Reduction of Alumina

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Abstract

This paper presents an updated calculation of the theoretical minimum energy required to electrochemically reduce alumina to aluminum using either consumable carbon anodes or oxygen evolving (non-consumable or inert) anodes. Knowledge of this energy (the standard enthalpy of reaction) is required for heat balance calculations. The reversible potential, or Nernst potential, needed to calculate cell voltage, is obtained from the standard Gibbs energy of reaction and the activities of reactants and products. A more conservative definition of energy efficiency is proposed. Problems involving heat balance and energy consumption when using oxygen-evolving anodes are discussed.

Definition of Terms

a .... Activity (thermodynamic concentration)

Bemf . Back electromotive force or counter electromotive force (V)

b ...... Mole or volume fraction of CO_2 reacting with carbon

CE .... Current efficiency (%)

E^o. ... Standard potential or standard emf (V)

E_{rev} .. Reversible or Nernst potential (V)

$E_{\Delta H}{}^o$ .. Aluminum producing voltage (V),

    $I(E_{\Delta H}{}^o)$ is the energy to make aluminum (kW)

E_{cl} .... Voltage within the heat loss boundary (V)

E_{cell} .. Total voltage of cell (V)

F ...... Faraday constant, 96.485 kJ /volt equivalent

G...... Boudouard reaction factor

ΔG^o ... Standard Gibbs energy of reaction (kJ/mol Al_2O_3)

ΔH^o .. Standard enthalpy of reaction (kJ/mol Al_2O_3)

I ...... Cell current or line current (kA)

n ..... Number of electrons transferred in the reaction as written

R_c ... Gas constant, 8.3144 J mol^{-1} K^{-1}

R .... Electrical resistance (ohm)

R_g ... Ratio CO/CO_2 of unburned cell gas

ROS Relative oxide saturation, ROS = %Al_2O_3 /Sat % Al_2O_3

T ..... Temperature of electrolyte (K)

T_e.... Temperature of electrolyte (°C)

T_r ... Ambient temperature (°C)

l ..... Liquid state

g ..... Gaseous state

s ..... Solid state

x ..... Fractional current efficiency, x = %CE / 100

y ..... Fraction α phase in alumina feed. y = % α /100

α .....Alpha crystalline state of alumina

γ ..... Gamma crystalline state of alumina

η_{sa}... Surface overvoltage at anode (V)

η_{ca}... Concentration overvoltage at anode (V)

η_{cc}.. Concentration overvoltage at cathode (V)

Subscripts:
    el ...... electrolyte
    bub .... bubbles
    an ..... anode
    ca ..... cathode
    ex ..... external

Introduction

Thermodynamic calculations are required to analyze energy consumption and cell voltage in aluminum production. The standard Gibbs energy of reaction, ΔG^o, and activity data are needed to calculate the Nernst potential, also called the

reversible potential. The standard enthalpy of the overall cell reaction is required to calculate the heat balance of the cell and to calculate energy efficiency.

The frequently used 1976 thermodynamic analysis by Bratland, Grjotheim and Krohn[1] is a bit out of date. We now have better thermochemical data. Moreover, their analysis was based upon alpha alumina. At that time most of the alumina used in Europe was floury alpha alumina, but today the cell feed is sandy, largely gamma alumina. Now we know that CO_2 is the primary anode product. CO is not made at the anode but by CO_2 (indirectly) re-oxidizing aluminum, and to a small extent by CO_2 reacting with carbon (the Boudouard reaction). Therefore, the standard Gibbs energy of reaction (used to calculate E^O) should be based upon CO_2 as the only gaseous product for carbon anodes and O_2 for non-consumable anodes. However, in determining the standard enthalpy of reaction, ΔH^O, for making an energy balance, the reactants are gamma alumina plus a small amount of alpha phase (and carbon with conventional anodes) entering at ambient temperature. The products are Al, CO and CO_2 (or Al and O_2 with non-consumable anodes) leaving at electrolyte temperature. This work supersedes such earlier calculations presented by Haupin[2] and Grjotheim and Kvande[3].

Cell Voltage

The voltage of a cell is the sum of the back emf (counter electromotive force), the electrolyte's ohmic voltage drop, the additional ohmic voltage drop in the electrolyte caused by anode gas bubbles, the ohmic voltage drops through the anode and cathode and voltage drops external to the cell.

$$E_{cell} = Bemf + IR_{el} + IR_{bub} + IR_{an} + IR_{ca} + IR_{ex} \qquad 1.$$

In heat balance calculations we are interested in the voltages within the heat boundary and omit the voltages external to the boundary. However, the external voltage drop must be included in calculating the energy consumption (kWh/kg Al) and in calculating the production costs ($/kg Al).

The back emf or counter emf is made up of the reversible potential plus overvoltages.

$$Bemf = E_{rev} + \eta_{sa} + \eta_{ca} + \eta_{cc} \qquad 2.$$

The reversible potential or Nernst potential is the voltage required to hold the cell in equilibrium. Knowing the cell reaction, the reversible potential can be calculated from the standard Gibbs energies and activities of the products and reactants.

The kinetics of producing products requires additional voltage at both electrodes (overvoltages). These overvoltages result from concentration gradients and the kinetics of surface reactions at the electrodes. They vary with electrolyte composition, current density and temperature and must be measured. Typical values are 0.47 V for carbon anode surface overvoltage, 0.03 V concentration overvoltage at the anode and 0.03 V for concentration overvoltage at the cathode. Oxygen evolving anodes have a combined concentration and surface overvoltage of about 0.1 V.
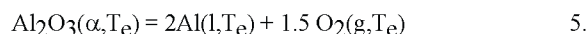
Reversible Potential

The standard potential, E^O, is based upon ΔG^O, the standard Gibbs energy change for the cell reaction with all reactants and products at electrolyte temperature and at unit activities:

$$E^O = - \Delta G^O/(nF) \qquad 3.$$

The cell reaction for Hall-Héroult cells with carbon anodes is:

$$Al_2O_3(\alpha, T_e) + 1.5C(s, T_e) = 2\,Al(l, T_e) + 1.5\,CO_2(g, T_e) \quad 4.$$
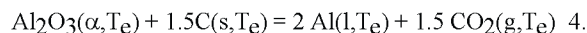
With oxygen evolving anodes, the reaction is:

$$Al_2O_3(\alpha, T_e) = 2Al(l, T_e) + 1.5\,O_2(g, T_e) \qquad 5.$$

Since the values of ΔG^O for the above cell reactions are positive, the E^O values are negative. Likewise, by thermodynamic convention, cell voltage and all components of it are negative because energy is consumed rather than produced. However, engineers and cell operators in the primary aluminum industry consider the cell voltage to be positive; hence in this paper, the sign of all cell voltage components will be made positive.

Alpha alumina was used in the cell reaction because gamma and other non-alpha aluminas convert to the alpha phase as they go into solution. There is strong evidence that at normal current densities the anode reaction with carbon anodes produces only CO_2.[4] When oxygen discharges onto carbon it forms a stable C-O surface compound.[5] At low oxygen evolution rates the surface compound can detach slowly from the surface and produce CO, but at normal current densities (high oxygen activity), additional oxygen atoms react with the CO surface compound before it can detach. This forms an unstable CO_2 surface compound that detaches rapidly as gaseous CO_2 and leaves fresh carbon for oxygen to deposit upon forming new C-O surface compound. Hence the anode product is CO_2.[4]

Most of the CO in the off gas is produced by a redox reaction between CO_2 and metal in the bath: either dissolved sodium, monovalent aluminum containing species, or impurities in the bath that were reduced to a lower valence state at the cathode. This results in a loss in current efficiency and produces one mole of CO for each mole of CO_2 reacting. A small additional amount of CO is produced by the Boudouard reaction whereby CO_2 reacts with carbon in the pores of the anode or with carbon dust in the bath, forming two moles of CO for each mole of CO_2 consumed. Substituting n = 6, F = 96,485 J / volt equivalent and numerical values for ΔG^O as a function of T (K), the standard potential for cells with carbon anodes (with its sign changed) becomes:

$$E^O = 1.898 - 0.0005733\,T \qquad 6.$$

For cells with oxygen evolving anodes:

$$E^O = 2.922 - 0.0005713\,T \qquad 7.$$

Note that the cell voltage would be about 1.03 V higher for inert anodes if the cell temperatures and overvoltages were the same. However, measurements indicate that the overvoltage on inert anodes is about 0.4 V less than on carbon anodes[6, 7]. If current

density and interelectrode spacing are not changed the net effect of inert anodes will be an increase in cell voltage of about 0.63 V. Adding a catalyst, such as ruthenium oxide, to the oxygen producing anode may save another 30 millivolts[7].

The Nernst equation corrects the standard potential for the actual activities to give the reversible potential, E_{rev}:

$$E_{rev} = E^o + \frac{R_cT}{nF} \ln \frac{a^2_{Al}\, a^{1.5}_{CO_2}}{a_{Al_2O_3}\, a^{1.5}_C} \qquad 8.$$

Al, CO_2, and C are present close enough to their standard states that they are assigned unit activities. The activity of alumina in the electrolyte must be obtained from measurements.

Activity of Alumina

New activity data for alumina have been obtained by Dewing and Thonstad[8] using cryoscopic data for the Al_2O_3–Na_3AlF_6 system and by Solheim and Sterten[9] using alumina concentration cell voltages for the Al_2O_3–Na_3AlF_6–AlF_3 system. Equation 9 provides a good fit to these data. Extrapolating to industrial compositions containing CaF_2, MgF_2 and LiF assumes that the effect of electrolyte composition on alumina activity is fully reflected in the alumina solubility at saturation.

$$a_{Al_2O_3} = -0.03791(ROS) + 2.364(ROS)^2 - 2.194(ROS)^3 + 0.868(ROS)^4 \qquad 9.$$

$$ROS = \%Al_2O_3/(sat\ \% Al_2O_3). \qquad 10.$$

At 960°C the reversible potential, E_{rev}, for cells with carbon anodes is 1.191V for electrolyte saturated with alumina. At 1/3 saturation E_{rev} is 1.222 V. For cells with oxygen evolving anodes, E_{rev} at 960°C is 2.217 V at saturation and 2.248 V at 1/3 saturation .

Voltage Equivalent of Energy to Make Aluminum, $E_{\Delta H}{}^o$

The term $E_{\Delta H}{}^o$ was previously called E_{Al} by Haupin.[2] Grjotheim and Kvande[3] did not convert ΔH^o to a voltage. Multiplying $E_{\Delta H}{}^o$ by cell current gives the part of the electrical energy input to the cell that is used in producing aluminum. The remaining energy becomes heat. Using $E_{\Delta H}{}^o$ avoids having to make a materials balance to calculate the heat balance.

In deriving $E_{\Delta H}{}^o$, the chosen heat loss boundaries were the steel shell of the cell, the top of the ore cover and the ore covered top and sides of the anode. $E_{\Delta H}{}^o$, the voltage equivalent of the total energy (enthalpy) to make aluminum becomes:

$$E_{\Delta H}{}^o = \Delta H^o\ (x)/(nF) \qquad 11.$$

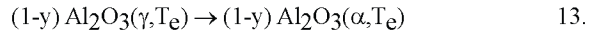where ΔH^o is the standard enthalpy of the cell reaction for reactants entering at ambient temperature and products leaving at electrolyte temperature. In the static state, the route from input to output need not be considered. Intermediate steps do not change

ΔH^o for the overall reaction. Current efficiency as a fraction, x, enters into the equation because only the aluminum actually made consumes energy.

The primary reaction in the cell is:

$$(1-y)Al_2O_3(\gamma,T_r) + y\ Al_2O_3(\alpha,T_r) + 1.5C(s,T_r) \rightarrow$$
$$2Al(l,T_e) + 1.5\ CO_2(g,T_e) \qquad 12.$$
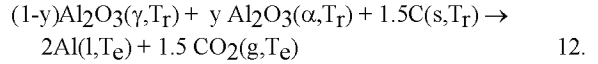
When the alumina contacts the bath:

$$(1-y)\ Al_2O_3(\gamma,T_e) \rightarrow (1-y)\ Al_2O_3(\alpha,T_e) \qquad 13.$$
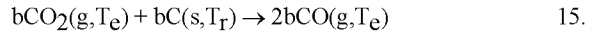
This reaction is spontaneous and slightly exothermic, but needs to be considered only in dynamic simulations.
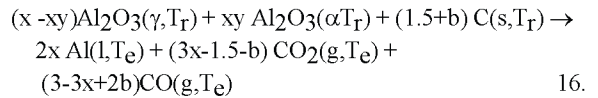
Some of the metal made at the cathode, through a series of reactions, dissolves into the electrolyte and back reacts with CO_2 made at the anode forming alumina and CO.

$$2(1-x)\ Al(l,T_e) + 3(1-x)\ CO_2(g,T_e) \rightarrow$$
$$(1-x)\ Al_2O_3(\alpha,T_e) + 3(1-x)\ CO(g,T_e) \qquad 14$$
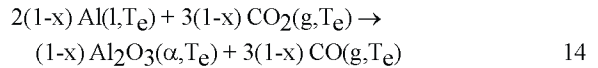
In addition to the CO_2 reacting with dissolved metal, a small fraction, b, reacts with carbon to make CO (the Boudouard reaction).

$$bCO_2(g,T_e) + bC(s,T_r) \rightarrow 2bCO(g,T_e) \qquad 15.$$

Adding reactions 12, 14 and 15 we get for the steady state:

$$(x-xy)Al_2O_3(\gamma,T_r) + xy\ Al_2O_3(\alpha T_r) + (1.5+b)\ C(s,T_r) \rightarrow$$
$$2x\ Al(l,T_e) + (3x-1.5-b)\ CO_2(g,T_e) +$$
$$(3-3x+2b)CO(g,T_e) \qquad 16.$$

Obtaining ΔH^o for reaction 16 allows one to calculate $E_{\Delta H}{}^o$ by equation 11.

The number of electrons transferred, n = 6 (x). Therefore,

$$E_{\Delta H}{}^o = \Delta H^o\ (x)\ /[6(x)F] = \Delta H^o\ /\ 6F \qquad 17.$$

Calculation of ΔH^o for the overall cell reaction, equation 16, requires knowledge of b. Its value can be calculated by equation 19, if one knows the current efficiency, x, and the CO/CO_2 ratio, R_g, for the unburned gas from the cell.

Equation 19 is obtained from equation 16 by evaluating the CO/CO_2 ratio, R_g:

$$R_g = (3-3x+2b)/(3x-1.5-b) \qquad 18.$$

Solving for b:

$$b = [3x\ R_g\ - 1.5R_g + 3x\ -3]/(2+R_g) \qquad 19.$$

The CO/CO_2 ratio, R_g, can be estimated using equations 20 and 21 from Beck[10].

$$\%CO = [200\ G - 2\ (\%CE) - 8]\ /G \qquad 20.$$

% CO_2 = 100 – %CO   21.

G ≅ 1.05 for cells with prebaked anodes

G ≅ 1.19 for cells with Soderberg anodes

Because Soderberg anodes are of poorer quality than prebaked anodes they produce more carbon dust, greater Boudouard reaction, a higher amount of CO in the unburned gas from the cell and, therefore, a higher G value. Indeed, the Boudouard reaction is about 20 times greater for Soderberg anodes than for prebaked anodes. By combining equations 19, 20 and 21 we can estimate b for prebaked anodes and Soderberg anodes.

For prebaked anodes:

$$b \cong -0.1082 + 0.1866(x) - 0.064(x)^2 \qquad 22.$$

For Soderberg anodes:

$$b \cong 0.0721 + 0.2233(x) - 0.079(x)^2 \qquad 23.$$

The CO/ CO_2 ratio can be obtained more accurately from chemical analysis of the unburned cell gas. Then equation 19 gives b directly.

There is research underway to develop an oxygen evolving (inert) anode. With such an anode, the cell reaction would be:
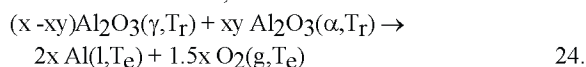
$$(x-xy)Al_2O_3(\gamma,T_r) + xy\ Al_2O_3(\alpha,T_r) \rightarrow$$
$$2x\ Al(l,T_e) + 1.5x\ O_2(g,T_e) \qquad 24.$$

Table I gives ΔH^o values for the cell reaction using gamma alumina (y=0), 25°C ambient temperature, several electrolyte temperatures, several current efficiencies and several values of b. Data are given also for inert anodes. JANAF table[11] values were used to make the calculations. To correct the ΔH^o values in table I for alumina containing some alpha phase add the correction factor Cor α (equation 25). To change to another ambient temperature add the correction factor Cor T_r (equation 26).

$$Cor\ \alpha = 18.83\ x\ y \qquad 25.$$

$$Cor\ T_r = (25 - T_r)\ [\ 0.0838\ (x - xy) + 0.080\ xy +$$
$$0.00871\ (1.5 + b)] \qquad 26.$$

The ΔH^o values in Table I were converted to voltages by dividing by 6F (578.91 kJ/ Volt equivalent) giving $E_{\Delta H}^o$ values per equation 17. Equations 27 and 28 were obtained by multiple regression analysis of these values (including the corrections for ambient temperature and for the alpha alumina in the cell feed). The electrolyte temperature, T_e, is in °C.

For carbon anodes:

$$E_{\Delta H}^o = 0.23706 + 4.6757e\text{-}4(T_e) - 2.25e\text{-}7(T_e)^2 +$$
$$x\ [1.4024 + 0.03253\ y + 2.23e\text{-}4(T_e)\ ] + b[0.3086 -$$
$$1.97e\text{-}5(T_e)] + (25\text{-}T_r)\ [0.000145(x - xy) +$$
$$0.000138\ xy + 0.000015\ (b + 1.5)] \qquad 27.$$

For inert anodes:

$$E_{\Delta H}^o = x[\ (2.881 + 0.03253\ y + 0.0002016\ T_e) +$$
$$(25\text{-}T_r)\ [0.0001448(1 - y) + 0.000138\ y] \qquad 28.$$

Equation 29 gives the required average rate of heat loss from the cell to maintain a constant temperature:

$$Heat\ loss\ (kW) = I\ (\ E_{cl} - E_{\Delta H}^o) \qquad 29.$$

Table I

100(x)=		$\Delta H°$ in kJ for 6 Faradays Current			
% CE	b	950 °C	960 °C	970 °C	980 °C
Enthalpy per reaction 16, no Boudouard reaction					
100	0.00000	1211.46	1212.94	1214.41	1215.89
95	0.00000	1164.73	1166.14	1167.55	1168.96
90	0.00000	1118.00	1119.35	1120.69	1122.04
85	0.00000	1071.27	1072.55	1073.83	1075.11
Enthalpy per reaction 16, prebaked anodes, Boudouard reaction					
100	0.01442	1214.12	1215.59	1217.07	1218.55
95	0.01136	1166.84	1168.25	1169.67	1171.08
90	0.00798	1119.47	1120.82	1122.17	1123.52
85	0.00421	1072.05	1073.33	1074.61	1075.89
Enthalpy per reaction 16, Soderberg anodes, Boudouard reaction					
100	0.21635	1251.38	1252.88	1254.38	1255.90
95	0.21288	1204.03	1205.46	1206.90	1208.33
90	0.20904	1156.59	1157.94	1159.31	1160.68
85	0.20478	1109.08	1110.39	1111.68	1112.98
Enthalpy per reaction 24, Inert anodes					
100	n.a.	1779.06	1780.23	1781.39	1782.56
95	n.a.	1690.11	1691.21	1692.32	1693.43
90	n.a.	1601.15	1602.20	1603.25	1604.30
85	n.a.	1512.20	1513.19	1514.18	1515.18

Components of Cell Voltage and Power

Typical values for the components of cell voltage and the distribution of power are summarized in Figures 1, 2 and 3. Figure 1 represents typical state-of-the-art technology for large (>300 kA) modern cells[12]. These plots allow comparison of the voltage determined by thermodynamics (reversible potential), by kinetics (overvoltages) and by ohmic resistance. The energy plots show the energy that goes into making aluminum and that which produces heat. $E_{\Delta H}°$ is larger than the reversible potential because some of the heat generated in the cell is used in the reduction of alumina. Comparing figures 1 and 2, note the increased heat loss caused by a lower current efficiency. Electrolyte temperature must rise to provide the greater heat loss and the higher temperature will cause further loss of current efficiency. Fortunately, melting of ledge moderates the rise in temperature.
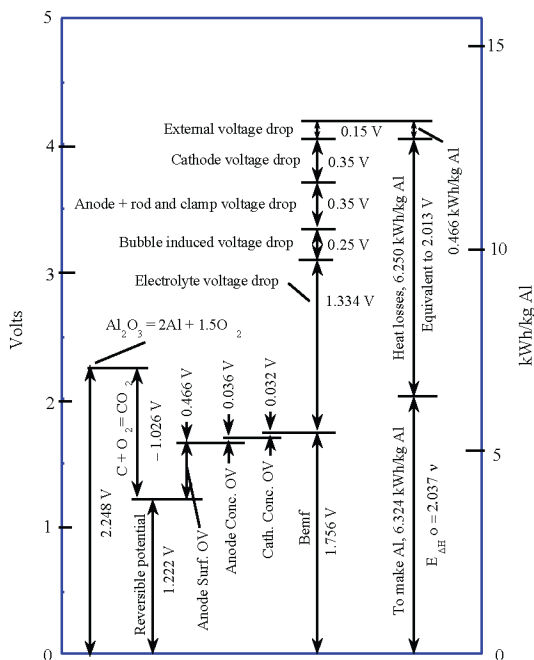
Fig. 1, Cell Voltage and Power @ 96 % CE, 4.19 V, 13.0 kWh/kg Al, 960°C, Alumina-10% Alpha-1/3 saturation, Carbon anodes
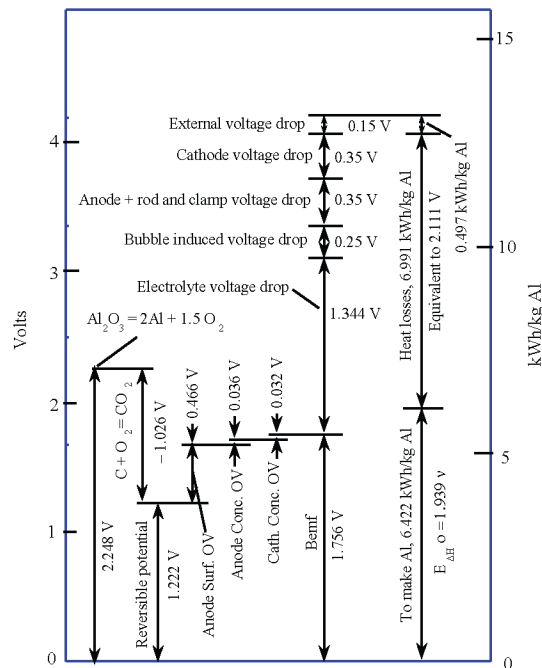

Fig. 2, Cell Voltage and Power @ 90 % CE, 4.20 V, 13.91 kWh/kg Al, 960°C, Alumina — 10% Alpha -1/3 saturation, Carbon anodes

Limitations

Equations 27 and 28 are valid for steady state operation. They neglect the following minor corrections. The first two add heat while the third removes heat. Heat is generated by aluminum fluoride (added at ~25 °C) neutralizing the soda content of the alumina. Each 0.1 % soda in the alumina lowers $E_{\Delta H°}$ by about 0.018%. Air burning of carbon anodes slightly raises their surface temperature thereby slightly lowering heat loss. Adding crushed bath extracts a small amount of heat.

In a dynamic energy balance, a running materials balance must be made. Introduction of each new anode and the heating, and dissolution of each alumina addition will extract heat from the electrolyte and cause transient drops in electrolyte temperature. The alumina will be heated somewhat in the ore hoppers on the cell and the conversion of γ alumina to α alumina as it enters the electrolyte will reduce slightly its cooling effect. The energy consumed by the reduction reaction will then be calculated for the reactants, alpha alumina and carbon (when using carbon anodes) at electrolyte temperature producing aluminum and gaseous product at electrolyte temperature.

Energy efficiency

Often energy efficiency is taken as the energy that goes into making aluminum at the existing current efficiency compared to the total electrical energy input to the cell:

$$\text{Energy Eff} = \frac{I\,E_{\Delta H}^{°}}{I\,E_{cell}} = \frac{E_{\Delta H}^{°}}{E_{cell}} \qquad 30.$$

This definition fails to penalize the energy efficiency value for the production of CO, a wasted potential source of energy. The authors believe that a better measure of energy efficiency is the ratio of theoretical kWh/kg Al at 100% current efficiency (no CO gas production) compared to the actual measured kWh/kg Al.

Energy Eff. = kWh/kg Al (theo) / kWh/kg Al (actual)   31.

For a carbon anode calculated by equation 12:

$$\text{kWh/kg Al (theo)} = 4.887 + 0.002058\,T_e - 6.71e\text{-}7\,T_e^{\,2} + 0.097\,y + 0.0005\,(25 - T_e) \qquad 32.$$

For an inert anode calculated by equation 24:

$$\text{kWh/kg Al (theo)} = 8.587 + 0.000601\,T_e + 0.097\,y + 0.0005\,(25 - T_e) \qquad 33.$$

Using equations 31, 32 and 33, cells operating at 960°C, 4.2 volts, 95% current efficiency, 25°C ambient temperature, with 10% alpha alumina feed, the energy efficiency would be 47.5%

for cells with carbon anodes and 69.6% for cells with inert anodes.

about 2.5% higher than that of Bratland et al. and finds the value is not a function of current efficiency. A more conservative definition is proposed for energy efficiency. If a switch is made to oxygen– evolving anodes, either a new more erosion resistant material will be needed for the cell's walls, or a cell design that packs much more production into a given volume.
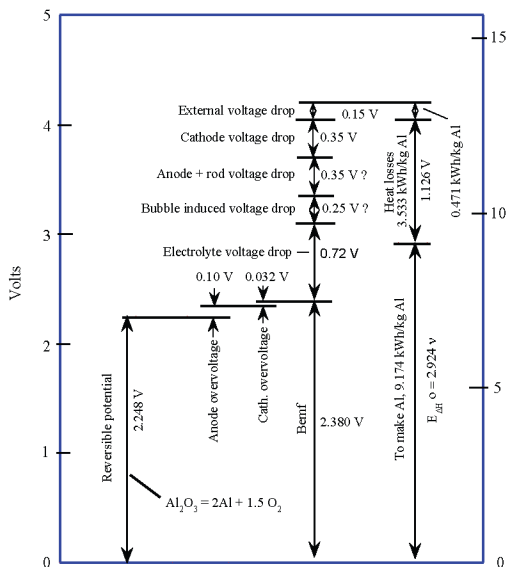
Fig. 3, Inert Anode Cell Voltage & Power Distribution @ 95% CE, 4.20 V, 13.2 kWh/kg Al 960°C, Alumina- 10% alpha-1/3 saturation, Inert anodes

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In order to operate cells with oxygen-producing anodes at the same voltage (same kWh/kg Al) as conventional cells, it will be necessary to compensate the higher Bemf by lowering the voltage drop through the electrolyte. This will require reducing the anode to cathode distance, and/or increasing electrode areas.

Because more of the electric power in these cells goes into making aluminum and less into producing heat, maintaining the electrolyte temperature will be difficult. Extra thermal insulation alone will not suffice. Present materials of construction require a protective layer of frozen electrolyte on the cell walls. Maintaining a frozen layer of bath requires a relatively large flow of heat through the walls (30 to 45% of the total heat loss). This so limits the ability to increase thermal insulation that a thermal balance will require more than just increased insulation. Ideally some new, unknown material resistant to attack by molten cryolite and molten aluminum, and also resistant to high temperature oxidation, will be needed for the internal walls of the cell. Lacking such a material, a means must be found to increase aluminum production (increase electric current) within the cell without increasing the current density. This, in principal, could increase heat generation sufficiently to supply the unavoidable heat losses.

Summary

New equations are presented to calculate that part of the electrical energy input to the cell used in producing aluminum. This energy is about 1% lower than that given by Bratland et al.[1] The rest of the energy produces heat, which must balance the heat losses from the cell. The new analysis gives a reversible potential