Influence of Melt Flow and Temperature on Erosion of Refractory and Deposit Formation in Induction Furnaces

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Abstract

Deposition and erosion phenomena in induction-channel furnace for Al melting in alumino-silicate refractory are considered. The possibility of simultaneous erosion and deposition in the same cross-section of the channel is shown by chemical reaction model, where the erosion of refractory is described by action of chemically active aluminium oxides and deposition by stable oxides. The variations of erosion and deposition in the same cross-section of the channel are explained by variation of the thickness of laminar sub-layer. A phenomenological model is described to study the stability conditions of a uniform growth of the build-up layer. Based on suitable approximations, analytical formulae have been derived enabling to evaluate the stability region in the concentration-temperature plane depending on the initial roughness of the growing surface.

1. Mechanism of Chemical Erosion and Deposition

The oxides with high melting point Al$_2$O$_3$, MgO and especially spinel structure Al$_2$MgO$_4$ are the dominating constituents in the deposits of Al melting channel furnaces. We will neglect the large variety of doping elements except Mg for a moment. Refractory material is build up with high silica fraction, because silica has the advantage of strong expansion effect for getting high material density after heating and for closing cracks after reheating [1]. However, chemical resistance of silica is lower than of alumina.

Al and Mg atoms are chemically very active and can easily take part in chemical reactions with refractory. The refractory contains such oxides as Fe$_2$O$_3$, P$_2$O$_5$, which are holding the oxygen atom less tightly than Al or Mg oxides. Hence, the reactions of Al and Mg with less stable oxides of refractory are possible. Al$_2$O$_3$ cannot be formed without intermediate stages, because the collision of several molecules is unlike. Moreover, Al$_2$O$_3$ formation at once cannot describe simultaneous erosion and deposition, because Al$_2$O$_3$ would deposit at the same point it was created. Hence, intermediate reaction products are necessary for modelling of the processes in the channel.

Consider that other aluminium oxides Al$_2$O, Al$_2$O$_2$ act as intermediate products, since they are less stable as Al$_2$O$_3$. Therefore, we will assume the following simple irreversible reaction scheme (see Fig. 1):

- reaction at refractory surface: Al$_2$O + refract. $\rightarrow$ Al$_2$O$_2$ +...
- reaction in bulk of Al melt: 2 Al$_2$O $\leftrightarrow$ Al$_2$O$_2$ + 2 Al
- reaction in bulk of Al melt: Al$_2$O + Al$_2$O$_2$ $\leftrightarrow$ Al$_2$O$_3$ + 2 Al
- deposition of Al$_2$O$_3$: Al$_2$O$_3$(in melt) $\leftrightarrow$ Al$_2$O$_3$(corundum)

The active Al$_2$O molecules are considered here to be responsible for the erosion of refractory. The reaction rates in bulk between oxides Al$_2$O, Al$_2$O$_2$, Al$_2$O$_3$ are low because they are reacting via collisions, and probability of such collisions is proportional to the product of respective densities. The formation of Al$_2$O occurs due to entropy factor. Al$_2$O$_2$
will be considered here as inert in respect to refractory, despite it is more active than Al$_2$O$_3$. Neither Al$_2$O nor Al$_2$O$_2$ form solid phase at 800 °C and so they do not make deposits. Al$_2$O attacks more chemically unstable parts into pores such as cement material that joining the grains. Consequently, the grains are able to liberate after sufficient amount of cement has been eroded. The erosion-deposition mechanism is shown schematically in Fig. 1. If the deposition rate is high enough, corundum layer delays or even blocks the access for Al$_2$O molecules in pores and erosion becomes impossible.

2. Erosion and Deposition Model

Let us consider the aluminium melt with small fraction of Al$_2$O, Al$_2$O$_2$ and Al$_2$O$_3$. The temperature is almost constant in channel cross-section that follows from characteristic turbulent velocities (about 1 m/s) and small channel diameter (10 - 20 cm) [2]. Calculations of melt dynamics in electromagnetic field showed that two vortices are characteristic in channel cross-section (see Fig. 2). The intersection of both vortices possesses a high turbulence with characteristic pulsation. The simultaneous erosion at points B, D and deposition at - A, C can be described by variation of thickness of laminar layer at refractory. High turbulence at points A, C significantly diminishes the thickness of laminar layer in comparison with points B, D.

Let us denote relative concentrations of Al$_2$O as $c_1$, Al$_2$O$_2$ - $c_2$, Al$_2$O$_3$ - $c_3$. The total concentration of oxygen is $c=c_1+2c_2+3c_3$. The minimum of free energy [3] at large enough Al$_2$O$_3$ concentration yields:

$$c_1 = c^{1/3} \exp \left[ \frac{\varepsilon_1 - \varepsilon_3 / 3}{kT} \right], \quad c_2 = c^{2/3} \exp \left[ \frac{\varepsilon_2 - 2 / 3 \varepsilon_3}{kT} \right], \quad c_1 \ll c, \quad c_2 \ll c. \quad (1)$$

There is still a significant number of active Al$_2$O and Al$_2$O$_2$ molecules because of entropy factor. Intense turbulence makes the distribution of oxides practically homogeneous in the bulk of channel cross-section. Variations occur only near refractory surface due to laminar layer. The concentrations of components $c_1$, $c_2$, $c_3$ vary according to diffusion equations, and $D_1$, $D_2$ and $D_3$ are diffusion coefficients for each component, respectively. Basing on diffusion-controlled assumption, the boundary concentration of Al$_2$O$_3$ at flat corundum deposit is equal to equilibrium concentration of Al$_2$O$_3$ in melt

$$c_{eq}(T) = A_{cor} \exp \left[ - \frac{E_{cor}}{kT} \right], \quad (2)$$

where the parameters $A_{cor} = 0.017$, $E_{cor} = 0.85$ eV are asymptotic values at 800 °C of Al-O phase diagram. The concentration of Al$_2$O at refractory surface is zero in diffusion-controlled approach. The laminar sub-layer at points A and C in Fig. 2 is the narrowest due to high turbulence. The laminar sub-layer at points B and D is much thicker.
The thickness of laminar layer \( \delta \) for turbulent flow and smooth refractory surface can be estimated as \([4]\)

\[
\delta = \frac{5\kappa}{u_*} = \frac{5\kappa |v_i v_j|}{u_*} \sqrt{\frac{3}{2}},
\]

where \( \kappa \) is kinematic viscosity coefficient, \( u_* \) is dynamic velocity that could be expressed by velocity pulsation, \( v_i \) are the velocity components. Coefficient \( \kappa \) is about \( 10^{-6} \) m\(^2\)/s for aluminium melt. Hydrodynamic simulations at points A, C (see Fig. 2) yielded \( u_* \approx 5 \) cm/s. Therefore, the thickness of laminar layer is \( \delta \approx 0.1 \) mm.

Fig. 2. Example of MHD flow in the cross-section of channel near inductor \([5]\). Model with two different thicknesses of laminar layer

Let us consider the case when the thicknesses of laminar layer are not constant along the channel perimeter as in Fig. 4. Let us assume that there are two characteristic thicknesses of laminar layer \( \delta_A, \delta_B \) and the sizes of both regions are equal as shown. The flux ratios in both regions are

\[
\frac{j_{1A}}{j_{3A}} = \left(1 + \frac{\delta_A}{\delta_B} \right) \left(1 + \frac{\delta_A + d}{D_1 + D_{1A}} \right), \quad \frac{j_{1B}}{j_{3B}} = \left(1 + \frac{\delta_B}{\delta_A} \right) \left(1 + \frac{\delta_B + d}{D_1 + D_{1B}} \right),
\]

where diffusion coefficient inside a pore \( D'_1 \) is lower than that in laminar layer \( D_1 \).

The dependence of fluxes in B is shown in Fig. 3 as a function of both ratio of diffusion coefficients inside the pores and ratio of the thickness of laminar layers. If laminar layer in B is thicker \( \delta_B > \delta_A \) at \( D'_1 = D'_{1B} \), one gets that the erosion dominates in B but deposition in A can be seen also in Fig. 3. The intensive deposition in region A reduces the diffusion of \( \text{Al}_2\text{O}_3 \) in pores. Due to stronger deposition, diffusion in pores is slowed down or cancels at all in this region, i.e., \( D'_1 > D'_{1A} \) \( = 0 \). Then follows that stationary flux ratio in B is

\[
\frac{j_{1B}}{j_{3B}} = 1 + \frac{\delta_B}{\delta_A}.
\]

This ratio can be much higher than 1. Henceforth, the strong erosion in B occurs and, consequently, increasing of the eccentricity of the elliptic cross-section. One can conclude that the intensity of erosion-deposition process increases during exploitation of furnace. The
resultant $\text{Al}_2\text{O}_3$ deposition flux towards the refractory at point A and $c_1 << 3$ $c_3 \equiv c_3 \equiv c_{eq}$ is

$$j_{3A} = \frac{1}{1 + \frac{\delta_A}{\delta_B} \frac{c_{eq}}{D_1 \cdot D'_{1A}}} \left( 3 \frac{c_{eq}}{kT} \right)^{1/3} \exp \left[ \frac{\varepsilon_1 - \varepsilon_{3} / 3}{kT} \right].$$

(6)

We can conclude that simultaneous erosion and deposition is describable by variation of the thickness of laminar layer. In order to decrease the rate of erosion, it necessary to improve the smoothness of the surface and increase the resistance of refractory to erosion active molecules. Making the thickness of laminar layer more homogeneous following from fluid dynamics calculations can essentially reduce the erosion rate. For this reason, chemical composition of more samples of various furnaces for aluminium melting should be performed.

3. Stability Conditions of Build-up Formation

We consider a simple model to study the stability of a uniform build-up formation. It is illustrated in Fig. 4. The surface $\xi = \xi(x, y, t)$ separates the solid build-up phase ($z < \xi$) from the molten metal ($z > \xi$). The liquid phase contains the build-up substance as an impurity with concentration $c(x, y, z)$ - the number of particles (molecules) per volume. It is assumed that the relative concentration (normalized to the concentration of basic substance particles) is small. The surface is flat except the region around $(x_0, y_0)$ where a half-spherical bulge of radius $r$ is located. The basic problem is to find the conditions at which the bulge develops or disappears during the process of impurity deposition. Our estimation is based on the ideas of the classical nucleation theory [6]. We assume that the rate of the process is limited by diffusion of the impurity through the concentration boundary layer of thickness $\delta(x, y)$. The liquid flow parallel to the surface is present, too. Both the concentration and the temperature are assumed to be constant, i.e., $c=c_0$, and $T=T_0$, outside of the concentration layer where the liquid flow is strongly turbulent. Similar model has been proposed earlier in [7], where the upper border of the concentration layer was assumed to be flat. Here we improve this model allowing that the upper border indicated in Fig. 4 by a dashed line is curved in general, as consistent with the real transport of fluid around the bulge. According to
this, we have \( \delta(x, y) = \delta_0 \) far away from the bulge and \( \delta(x_0, y_0) = \delta(r) \) on its top. We assume the linear approximation

\[
\delta(r) = \delta_0 \left(1 - \beta \cdot r / \delta_0 \right),
\]

where \( 0 < \beta < 1 \) is a coefficient which characterizes the relative decrease of the thickness \( \delta(x, y) \) of the concentration layer over the bulge of radius \( r = \delta_0 \). Such a linearization makes sense if the relative variation \( \beta \cdot r / \delta_0 \) is not large. Our previous approximation \[[7]\] is recovered at \( \beta = 1 \). We allow in general that the coefficient \( \beta \) can depend on the velocity of fluid along the surface, as well as on other parameters of the model, except \( r \). One may expect that a bulge of size \( r \sim \delta_0 \) will smoothly curve a relatively slow flow of molten metal around it, practically not changing the thickness of the concentration layer. It means that \( \beta(v) \approx 0 \) holds at small velocities \( v \) of a steady flow along the surface. If the flow velocity is large, or if the flow strongly fluctuates in such a way that \( v \) sometimes reaches relatively large absolute values, then the concentration layer on the top of a bulge of size \( r \sim \delta_0 \) can be destroyed, which corresponds to \( \beta \approx 1 \). The bulge tends to dissolve if \( r < r_{\text{crit}} \), where \( r_{\text{crit}} \) is the critical radius. If larger bulges are present, they tend to develop, which causes the instability of flat depositing surface and formation of porous or dendrite-like build-up layer. The critical radius is a solution of equation (24) in \[6\]. Including the factor \( \beta \), it reads

\[
\frac{\beta r_{\text{crit}}}{\delta_0} = \frac{bs}{bs + [DkT \cdot \Delta c / c_{eq}][2ae \cdot v_i \cdot (1 - l/l) \cdot (1 + c_{eq} / \Delta c)]},
\]

where \( \Delta c = c_{eq} - c_{eq} \) is the supersaturation, \( e \) is the binding energy and \( a^3 \) is the volume per one particle (molecule) in the deposited solid substance, \( l \) and \( l_i \) are the number of nearest impurity particles in the bulk of solid and on the depositing surface, respectively, and \( b \) is a phenomenological constant of order 1 introduced in \[7\]. Taking into account that \( Q >> 1 \) holds in a typical case of the induction channel furnaces (where \( Q \) is of order \( 10^5 \) or even larger), we obtain an approximate solution of (8)

\[
\frac{\beta r_{\text{crit}}}{\delta_0} \approx \frac{bs}{bs + [DkT \cdot \Delta c / c_{eq}][2ae \cdot v_i \cdot (1 - l/l) \cdot (1 + c_{eq} / \Delta c)]},
\]

where \( v_i \) is the velocity at the border of the laminar sublayer of thickness \( \delta_i \) and \( s = \delta_0 / \delta_i \). An approximation \( dv / dz \approx v_i / \delta_i \) has been used. Our approximations are valid if \( \beta r_{\text{crit}} / \delta_0 \) is remarkably smaller than \( \delta_i \) and \( \delta_0 \). To estimate the order of magnitude of \( \beta r_{\text{crit}} / \delta_0 \) expected in our application, we have set \( bs = 1, v_i = 10 \text{cm/s}, D = 10^{-5} \text{cm}^2 / \text{s} \), and \( l_i / l = 1 / 3 \). This yields at \( T = 1600^\circ \text{C} \):

\[
\frac{\beta r_{\text{crit}}}{\delta_0} \approx \frac{1}{1 + g_i(\Delta c / c_{eq})(1 + c_{eq} / \Delta c)} \quad \text{for Al}_2\text{O}_3 \quad (c_{eq}/c_i \approx 0.00092)
\]

\( g_i = 3.07 \), for \( MgO \) \((c_{eq}/c_i \approx 0.00692)\) - \( g_i = 5.68 \). In Fig. 5 we have shown the corresponding stability region in the \( \beta r / \delta_0 \) and concentration (the content in mass percents) plane at two different temperatures - \( 1550^\circ \text{C} \) and \( 1800^\circ \text{C} \).
Equation (9) explains the empirically observed different regimes of the build-up formation: the growth of dense layer with smooth surface characterized by the initial roughness-parameter \( r < r_{crit} \), and the growth of porous build-up layer which most often is observed in region A (Fig.5), where the stability condition \( r < r_{crit} \) is violated because of too small value of the critical radius \( r_{crit} \). The latter is consistent with a relatively smaller thickness of the concentration layer \( \delta_0 \), as well as a larger value of the parameter \( \beta \) due to relatively strong fluctuations of the flow velocity near the surface.

Conclusions

The deposition-erosion model is made which shows the mechanism of simultaneous erosion and deposition in the same channel cross-section of Al melting furnace. The necessary multistage oxidation scheme is constructed which includes aluminium oxides acting as intermediate products.

Based on suitable approximations, analytical formulae have been derived enabling to evaluate the stability region in the concentration-temperature plane depending on the initial roughness of the growing surface.

References


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Fig. 5. Stability and unstability regions for the growth of a flat build-up layer depending on the radius \( r \) and the concentration in mass percents (solid line \( Al_2O_3 \), dashed line \( MgO \)).