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A FINITE ELEMENT COMPUTATIONAL FLUID DYNAMICS SENSITIVITY ANALYSIS FOR THE CONCEPTUAL DESIGN OF A CARBOTHERMIC ALUMINIUM REACTOR

Dimitrios I. Gerogiorgis and B. Erik Ydstie
Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

Abstract
The inherent complexity of numerous industrial processes in the metallurgical industry poses formidable technical challenges for modeling, design and simulation: consequently, it is exceptionally difficult not only to optimize the operation of traditional plants without extensive experimentation, but also to model and analyze the feasibility and profitability of various novel designs proposed. Most metallurgical unit operations encompass a wide spectrum of phenomena (convection, diffusion, reaction, external field effects) that often occur simultaneously in multiphase configurations. Accurate modeling of complex distributed chemical processes entails the use of partial differential equation (PDE) descriptions which can now be routinely handled using commercial solvers; an attractive feature of the latter is a potential to handle combinations of diverse physical phenomena that occur in complex processes and yield highly coupled nonlinear PDE mathematical models. Thus, detailed steady state simulations of metallurgical processes can be obtained via advanced interactive software environments. A steady state sensitivity analysis of state variable distributions (namely, potential, absolute temperature and molten slag velocity) with respect to a key design variable (imposed electrode voltage) is performed for a conceptual carbothermic reduction reactor proposed by Johansen et al. (2000) for production of aluminium. Simultaneous solution of charge, heat and momentum balances on a suitably simplified two-dimensional computational domain representing a section of this reactor is used to achieve the goal of this work, which is to study trends important in efficient design. The major conclusion of the present CFD sensitivity study is that the voltage imposed on the horizontal heating electrode pairs affects the interplay between heat conduction and convection, thus the topography and uniformity of the temperature distribution. The electrode voltage has been qualitatively proved to govern the location and size of high-temperature regions in the reactor, thus affecting the advance of the endothermic carbothermic reduction reaction as well as the per volume reactor productivity. Therefore, electrode voltage is a crucial reactor design parameter that can also be used very conveniently as a manipulation variable for the efficient operation and control of a carbothermic reactor.

Introduction
The aluminium industry is recognized as one of the strongest, most capital- and energy-intensive industries of the twentieth century, literally shaping technology in modern times. Aluminium is the most abundant metal in Earth’s crust (8%); nevertheless, most processes developed for obtaining metallic aluminium from its various compounds are generally very far from economical [1]. Ever since Charles Hall invented electrolysis of alumina in a fused cryolite bath in Oberlin, OH (USA) and Paul Héroult was granted a patent for a similar process in Paris, France (both in 1886), the high-temperature, fluoride-assisted electrolysis of molten alumina for the production of pure aluminium remains unchanged in principle, despite many important technological modifications. The Hall-Héroult electrolytic aluminium oxide reduction process is recognized as the only technology for aluminium production that is both technically feasible and economically profitable [1]. Nonetheless, electrolysis of aluminium oxide is certainly not an environmentally benign process, since it is very energy-intensive and rapidly depletes natural resources for the production of the enormous electric energy required for electrolysis of alumina [2]. Extensive use and inevitable emission of cryolite bath fluorides also creates environmental problems and raises relevant issues [3]. The direct chemical reduction pathway has been identified long ago as a less expensive and more benign process alternative that can appreciably improve total volumetric reactor productivity [4]. The importance of reducing the historically consistent sensitivity of the aluminium industry to energy prices is another major reason to invest in research and development of potentially applicable as well as profitable process alternatives for aluminium production. The significance of this incentive is clearly illustrated by the impact of a recent energy crisis on aluminium production levels: in fact, the average monthly U.S. primary aluminium production plummeted by more than 28% between the years 2001 and 2002, as a result of the major energy crisis that struck the United States in early 2001, inducing major disruptions in production plants [5]. Aluminium industries have thus championed the development of carbothermic processes, particularly during energy crisis periods; this explains why investigation begun several decades ago [6].

Reynolds Metals Co. was among the pioneers of research efforts, with an extensive experimental and modeling program undertaken at their Alabama laboratory facility between the years 1971–1990. This detailed investigation resulted in several technical reports and a number of publications summarizing these efforts [7, 8, 9]; it also identified important technical challenges (showstoppers) that are to be overcome to develop a viable and profitable process. Thus, vaporization of gaseous aluminium species (Al_{2}O_{3}, AlO) produced by superheating at the high reaction temperatures made it clear that their recovery and recycling are elemental to viability. Reaction staging in distinctly different, separate reactor sections also emerged as a necessity for improving process controllability in most of the proposed reactors [4], since significant technical obstacles have been anticipated to hinder the transition from a laboratory scale reactor to a scaled-up industrial reactor prototype.

Kusik et al. of Arthur D. Little, Inc. later published the main conclusions of a thorough technoeconomic assessment study [10], based on a flowsheet for carbothermic reduction developed by a technical committee for the U.S. Department of Energy. Therein, the issue of individual technical challenges is illustrated again: the remark that “several of the subprocesses in the flowsheet have not been reduced to practice, and an opportunity exists to further improve process economics through research and development” clearly encouraged further pursuit of this ambitious endeavor. ALCOA Inc. thus spearheaded research in the field in later years; the initiated collaboration with ELKEM ASA Research (Norway) led to a multidisciplinary initiative upon which our study is based.
Carbothermic Reduction Technology

Carbothermic reduction is the only non-electrochemical process that has been proposed and tested for aluminium production [2]. The high reaction temperatures entail the use of electric energy for heating, but a carbothermic process is more energy efficient and demonstrates remarkably high theoretical volumetric productivity. Thus, considerable industrial reactor advantages can be achieved and significant economies of scale can be realized in production. The interest in carbothermic reactor technology is hence driven by significant cost advantages addressing the two largest cost sectors: (a) an identified potential for capital investment reduction due to higher reactor productivity and lower maintenance requirements than those characterizing conventional electrochemical processes. (b) a significant potential for higher energy efficiency which can be achieved by utilizing carbon and carbon monoxide off-gases (as useful energy sources for electric power cogeneration) and by the efficient management of thermal energy via integrated design.

Nevertheless, carbothermic reduction of alumina to aluminium is also a remarkably complicated process for many different reasons: (a) Aluminium carbide (Al4C3) formation is thermodynamically favored at temperatures below aluminium formation and becomes a key intermediate species that is inevitably affecting process balances, chemical equilibrium and overall conversion [2-4, 9]. (b) The process temperatures are required to be extremely high (exceeding 1900 °C in melting and 2100 °C in reduction zones), resulting in significant radiation heat losses for most proposed reactor configurations; heat management challenge is thus equally important for technical feasibility and for economic viability [2]. (c) Carbon monoxide (CO) is the overall gaseous product, leading to a high greenhouse penalty and raising environmental concerns. Nonetheless, energy recovery is possible by heat exchange and subsequent gas turbine combustion for power cogeneration [2, 4]. (d) Aluminium has a significant vapor pressure and evaporates readily at the reaction temperatures and at atmospheric pressure. This is aggravated by a tendency for vapor aluminium suboxide (Al2O) formation. Both problems can be addressed by alloy formation or by Al/Al2O vapor recovery towards Al4C3 formation. Carbon has been proposed as a reducing agent, since a plethora of experimental studies have documented that even some relatively low-purity forms can effectively advance the reduction [8, 9]. Carbothermic reduction of alumina to aluminium is extensively investigated: the literature survey reveals numerous studies that were conducted at laboratory and pilot-plant scale [4, 7-9, 11-13]. Several process pathways and reactor configurations have been proposed and tested unsuccessfully for commercialization [4], since the implementation of such proposals to the industrial scale has been hindered by vast complexity and limited controllability.

Carbothermic reduction can be a two-stage reaction process [14]. This is a complicated, reversible chemical reaction phenomenon with multiple species and phases involved in its advance; although species identification has been conducted, the actual reaction mechanism and a rate model have not been elucidated yet [2, 4]. The main reactants are aluminium oxide (Al2O3) and carbon (C), the key reaction intermediates are aluminium suboxide (Al2O) and aluminium carbide (Al4C3), and the reaction products are molten aluminium (Al) as well as carbon monoxide (CO) (a by-product). The complexity of the process is illustrated by the simultaneous coexistence of solids (C, Al2O3, Al4C3) and gases (CO, Al, Al2O) in the multicomponent molten slag that is formed in the reactor; this, in itself, explains many of the pressing modeling challenges.

Carbothermic Reactor Engineering

A wide variety of industrial scale process and reactor designs have been proposed in the literature over the years [4]; the complexity has evolved with the advances in understanding complex physics. This study is focusing on a carbothermic aluminium reactor patent of Johansen and Aune assigned to ALCOA and ELKEM [15]; the detailed schematic of the proposed reactor is provided in Figure 1. A clear observation is the effort to take advantage of chemistry by separating process phenomena in the following distinct stages.

1. The first stage of the process is a pre-reduction smelting zone. Carbon and aluminium oxide pellets are continuously fed to the submerged arc smelter, melt and react to form a viscous binary molten slag, contained in an inert-atmosphere, oil-cooled reactor. The reaction of aluminium oxide with an excess of carbon to form the Al4C3-rich slag of the first stage is written as (T > 1900 °C):

   \[ 2\text{Al}_2\text{O}_3(s) + 9\text{C(s)} \rightarrow (\text{Al}_4\text{C}_3 + \text{Al}_2\text{O}_3)_{\text{slag}} + 6\text{CO(g)} \]  
   \[(1)\]

2. The second stage is the high-temperature reduction zone: the first-stage molten slag flows slowly into the actual multi-electrode submerged arc reactor, where it is heated to a higher temperature, avoiding local surface superheating caused in open arc reactors. Liquid Al droplets and CO bubbles are rapidly generated at hot spots, while the chemical equilibrium can be assisted by further Al4C3 injection from the third stage, to avoid carbon depletion (if free C is exhausted, the reduction advance becomes problematic, promoting multiphase flow and simultaneous foaming effects [2]). The decomposition of the Al4C3-rich slag of the first stage to form the Al-rich phase of the second stage is written as (T > 2000 °C):

   \[ (\text{Al}_4\text{C}_3 + \text{Al}_2\text{O}_3)_{\text{slag}} \rightarrow (6\text{Al} + \text{Al}_2\text{C}_3)_{\text{metal}} + 3\text{CO(g)} \]  
   \[(2)\]

3. The third stage consists of a vapor recovery reactor (VRR), where Al and Al2O vapors react with C to form Al4C3 [16]. Vaporization occurs as CO vapors sweep the second stage reactor; unless Al species are recovered countercurrent to incoming solid feed, metal loss has a catastrophic impact on process economics, since it is shifting the equilibrium and sharply decreasing yield. This undesirable vaporization effect is reduced by staging and feeding the first and second stage gas streams to the VRR stage. The recovered Al4C3 (recycle stream) is reinjected into the reactor, minimizing metal vapor emission and maximizing process yield. Countercurrent flow exceeds incoming reactant preheating needs, allowing for energy recovery via heat exchange and cogeneration.

4. The fourth (final) stage of the process is the purification zone: liquid aluminium (of lower density than the slag) produced in the second stage floats and flows through an overflow weir to a tank. The latter serves as a molten metal flotation and skimming unit, where entrained solid Al2O3 particles and dissolved C material can be removed by proprietary technology to recover pure aluminium.

Although the theoretical foundation of process chemistry is sound, implementation details set the stage for the feasibility challenges. These are due to the technical difficulties associated with handling multiphase molten slags and to the extremely high temperatures (> 2000 °C) required for this reversible endothermic reduction. Simultaneous production of Al and Al2O vapors is also inevitable in any high-temperature reactor due to localized superheating, while the coexistence of molten slag, Al2O3 and Al4C3 particles is governed by thermodynamic equilibrium (temperature profiles). Any excess of Al2O3 may lead to aluminium oxy carbide (Al2O3C) as specified by the pseudobinary phase diagram of the system [4].
Carbothermic Reduction Process Modeling

Process modeling was recognized as an indispensable element for research and development of industrial carbothermic processes, leading to a remarkably thorough modeling study early on [7]. Nevertheless, modeling is itself hampered by numerous problems. The thermophysical properties of the system are not documented adequately in the range of high temperatures encountered [17-21]. The extreme structural complexity of the proposed reactor renders the design effort unique and far less straightforward. Unlike most widespread chemical processes that feature a certain degree of structure that is oriented to sharp staging of phenomena (e.g., mixing, reaction, separation, purification), this reactor draws its fundamental advantage (volumetric productivity improvement) by combining (rather than separating) phenomena in the second stage, where the slag decomposes to yield molten aluminium. Also, unlike definitive dimensional characteristics of conventional unit operations (height, diameter, packing, equilibrium stages), the geometry and components of the proposed carbothermic reactor will have to be customized in a much more elaborate fashion, rendering the design effort unique and far less straightforward. Accordingly, the carbothermic aluminium reactor would be much less open to standard engineering simplifications (perfect mixing, uniform flow profiles, or convection vs. diffusion domination), exactly because a plethora of phenomena occur simultaneously therein, and it is not clear which are dominant in different regions.

Experimentation, especially water modeling techniques [28-31] resolve similar questions (for a variety of metallurgical processes which comprise advanced multiphase high-temperature reactors) and facilitate greatly sound approximations for reliable models. Nevertheless, since most thermophysical properties of the slag are unknown, water modeling is not a suitable option at this stage (as thermophysical and transport properties have not been elucidated). Consequently, it is very risky to oversimplify a slag flow model, even when not considering multiphase flow due to gas generation.

The complex molten slag flow induces complex process dynamics and the foregoing discussion makes clear that a reliable chemical process ensuring operability and controllability must take into account a detailed PDE flow model of the second reactor stage. Without the latter, it would be questionable to postulate which regions within the reactor are diffusion- or convection-dominated. A flow model should ultimately quantify gas generation and metal production, to assess reactor productivity and process viability. Diffusion is conceptually promoting the microscale phenomena occurring in the reactor (melting, dissolving, component diffusion) while assisting the chemical reaction system reach equilibrium. Nonetheless, dominance of diffusion in extended reactor regions does not prevent the development of thick heat boundary layers (which can be induced by electrothermic superheating and in turn sustain high and potentially catastrophic temperature gradients).

Convection is arguably facilitating the circulation of molten slag, enhancing macroscopic transport of reactive slag and products. Nevertheless, intense recirculation can certainly be problematic, especially when resulting in high Reynolds and Peclet numbers: brief residence reduces conversion and compromises productivity. Several convective flow types are identified in the second stage: forced convection due to the incoming flow from the first stage, natural convection due to the temperature gradients developed, and dispersed convection due to the formation of CO gas bubbles. A flow model must ideally elucidate the interplay of phenomena.
Computational Fluid Dynamics Model

The second-stage submerged multielectrode carbothermic reactor is modeled employing the symmetry of design, considering only a two-dimensional section perpendicular to the major reactor axis at a plane defined by the horizontal electrode circular tip (Figure 1). The resulting two-dimensional computational domain comprises a narrow underflow inlet duct feeding the first-stage molten slag and the expansion to the first portion of the second-stage reactor with the tip of the inert graphite electrode immersed in the slag.

The full reactor and homogeneous slag assumptions are necessary in order to simplify the complex hierarchy of physical phenomena and first study the electric charge, heat and momentum balances. Thus, the goal here is to solve the steady state PDE problems for the respective variables of the latter balances (potential (V), field intensity (E), temperature (T), velocity (Ux, Uy), pressure (P)), obtaining reliable starting points so as to solve molar balances for species concentration profiles and reactor performance evaluation. Constant thermophysical properties are assumed throughout this study, although a temperature-dependent electric conductivity has also been used previously to illustrate the strong coupling between the charge balance and a Joule heat generation term [32].

Incompressible flow without buoyancy momentum generation is assumed, and model parameters have already been published [32].

The finite element method is used to formulate the PDE problem on a fine, unstructured triangular domain discretization (Figure 1). The developed finite element model of the reactor has been solved with quadratic finite element basis functions, using a commercial finite element simulation environment (FEMLAB® v. 2.3) [33]; three FEMLAB® modules have been used (Conductive Media DC, Incompressible Navier-Stokes, Convection and Conduction). A stepwise procedure (one more PDE problem solved at a time) can be initially followed so as to handle strong balance couplings, but the crucial goal considered during this study is the possibility of routine simultaneous solution of all three balances of the model (since the latter is to be used as an efficient process design tool).

CFD Equations and Boundary Conditions

Electrothermic convection modeling yields a triple-PDE problem. The first part of the CFD model is the steady state charge balance:

\[
(2D \text{ Laplace Equation}) \quad \nabla^2 V = V_{xx} + V_{yy} = 0 \tag{3}
\]

The second part of this model is the steady state heat balance:

\[
(2D \text{ Heat Equation}) \quad \nabla \cdot (kVT - \rho C_p TU) + \sigma VV^2 = 0 \tag{4}
\]

The third part of the model is the steady state momentum balance:

\[
(2D \text{ Navier-Stokes Equation}) \quad \rho(U \cdot \nabla U) - \mu(U^2 U) + \nabla P = 0 \tag{5}
\]

The boundary condition around the tip is the imposed voltage (V0) and a zero voltage (not a standard zero gradient) is used on the reactor inlet and on all reactor walls for the electric slag insulation (a zero gradient symmetry condition is used on the reactor outlet). The inlet temperature of slag (2173 K) and cooled walls (473 K) is set; zero gradient (heat insulation) is assumed at electrode tip. The symmetry boundary condition is again used on reactor outlet. A plug flow inlet velocity profile is considered (U0 = 0.001 m.s⁻¹) with a no-slip boundary condition on electrode tip and all walls; a zero pressure boundary condition is assumed on the reactor outlet.

The electrode characteristics are variables of extreme importance, governing field intensity and electric current density distribution; electrodes are also rigid bodies immersed into the reactive slag, perpendicular to the principal flow direction: convection patterns and steady state profiles are obviously affected by their presence. Electrode voltage and spacing are crucial, because they affect the field intensity distribution, hence the Joule effect heat production. This in turn is expected to affect the uniformity of heat generation, thus the uniformity of temperature and concentration distributions. Consequently, since voltage is a convenient manipulated variable, it is vital to study and understand its individual effect by a sensitivity analysis conducted for two voltage values (50, 100 V). The simulation results obtained are presented in Figures 2 and 3.

Figure 1: Schematic of the proposed carbothermic aluminium reactor [15] and the corresponding two-dimensional computational domain.
Figure 2: Two-dimensional distributions obtained from the CFD sensitivity analysis simulations for the electrothermic effect in the reactor: (a) Electric potential distribution, $V$, (b) Field intensity distribution, $E$, (c) Current density vector field, $J$, (d) Temperature distribution, $T$. 

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Figure 3: Two-dimensional distributions obtained from the CFD sensitivity analysis simulations for the electrothermic effect in the reactor: (a) Superposition of flow on temperature map, (b) Velocity distribution, U; (c) Velocity contour profile, U; (d) Pressure contour profile, P.
The present study focuses on the CFD modeling of a conceptual carbothermic reactor proposed for aluminum production and on a concise sensitivity analysis performed for two parameter values. A detailed, first principle, steady state finite element CFD model of the second stage reactor has been developed and used for the simultaneous solution of charge, heat and momentum balances on a two-dimensional computational domain representing the inlet underflow duct and the first reactor portion receiving molten slag. The potential, temperature and velocity distributions obtained by the simultaneous solution of charge, heat and momentum balances for the two values of the sensitivity analysis variable (voltage) reveal several trends that must be considered for process design.

The use of quadratic finite element basis functions has been proved particularly successful for the purpose of our simulations; the standard nonlinear iterative solver provided in FEMLAB® 2.3 (using the GMRES method and Incomplete LU preconditioning) has been used effectively to conduct the computational analysis. Sensitivity calculations can either be performed sequentially or, more efficiently, using a parametric solver as an extra component. The standard gradual mesh refinement and reinitialization strategy facilitates convergence, and choice of initial values is not critical. Nevertheless, reduction of local error is a challenge for the area close to the acute point formed at the top portion of the inlet duct. The CPU time required for convergence below tolerance ($10^{-15}$) when solving the triple PDE problem for a specific voltage value ranges from 5 minutes (586 triangles) to 1 hour (10499 triangles). Solutions are obtained using reasonable computational resources (a Pentium III / 1.2 GHz with 512 MB of RAM was used here).

The electric potential ($V$) distribution is depicted in Figure 2 (a) for the two different values of the imposed electrode voltage ($V_0$). The form of the solution is of course identical for both values, and confirms the suitability of the zero voltage boundary conditions used, since the profiles are exact two-dimensional projections of the three-dimensional solutions obtained in a previous study [32]. The potential contours are concentric ellipsoidal open curves that are unaffected by the zero voltage boundary condition at the inlet. The saddle point of the profile in the middle of the outlet edge is a result of the symmetry boundary condition that is imposed therein.

The field intensity ($E$) distribution is depicted in Figure 2 (b) and clearly indicates appreciable potential gradient values ($E = -\nabla V$) which are only encountered within a small radius from the circular edge of the electrode that is used for heating in the second stage. Intensity rapidly diminishes as one moves towards reactor walls: this obviously affects the extent of Joule heat generation greatly, since the Joule effect production term included in the heat balance is proportional to the square of the field intensity [$Q_J = \sigma (VV')^2$]. Consequently, conductive heat flux induced by the Joule effect is expected to be important only considerably close to the electrodes.

The temperature ($T$) distribution is depicted in Figure 2 (d) and reveals three distinct zones clearly affected by electrode voltage: the primary high-temperature zone at the bottom of the reactor, the secondary uniform upper zone upstream from the electrode and the superheated upper zone downstream from the electrode. The first occurs due to the significant convective heat flux at inlet, the second is explained by the local recirculation flow profile and the third is a clear result of the localized Joule heat generation. Consequently, convective heat transport prevails in the first two regions, while conductive heat transport is dominant in the third.

The sensitivity analysis reveals another important conclusion: although temperature plots are clearly different for the two values of the imposed voltage, the velocity contour plots are identical. This is due to the weak coupling of heat and momentum balances, in contrast to a Joule coupling between charge and heat balances. Electrode voltage emerges as an effective manipulation variable for reactor core temperature control, but not for slag solidification near reactor walls, as its effect is strong only in a limited region. The value range explored (50-100 V) is wide enough for design.

The triple-PDE CFD model used in this work is to be expanded to include a more elaborate description of physics in the reactor, since its present version is mainly addressed at probing the interplay between Joule heat generation and forced convection, providing upper bound estimates for electrothermic phenomena. Considering a vertical natural recirculation flow resulting from high temperature gradients via a Boussinesq approximation [27] is elemental to reliable reactor modeling, despite the parametric uncertainty associated with the thermal expansion coefficient ($\beta$). Nonetheless, the natural convection is expected to be a lot more important in the vertical plane perpendicular to the incoming flow.

Performing a sensitivity analysis for various reactor dimensions is a priority of paramount importance to reactor development, as it can directly answer “what-if” questions and foster modifications. A more accurate CFD sensitivity analysis study will entail use of a full thermodynamic model for reaction heat consumption [24] and temperature-dependent thermophysical property models [32]. Our model implementation can readily accommodate improved models as property measurements become available: this is not only an advantage, but also a necessity so as to achieve validation.

Finally, and because the carbothermic reduction reactor analyzed here is an excellent example of a complex process relying on many different and interdependent physicochemical phenomena, this CFD sensitivity analysis serves as a foundation for our current multiscale modeling for process design and control [32, 34-38].

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References

4. K. Motzfeldt et al., Carbothermic Production of Aluminium – Chemistry and Technology (Düsseldorf: Alum. Verlag, 1989).
30. Int. J.