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PROCESS SYSTEMS TOOLS FOR DESIGN AND OPTIMIZATION OF CARBOOTHERMIC REDUCTION PROCESSES

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Abstract

Collaborations between metallurgical industries and universities can be advantageous to both and evolve into win-win situations, because carefully planned and jointly executed research programs can benefit corporate research and development (R&D) and simultaneously promote basic and applied academic research. Successful partnerships provide exciting opportunities, far more interesting than mere technical consulting or abstract endeavors without application focus.

This paper presents the story of a fruitful collaboration among ALCOA Inc., (world leader in primary aluminium production), ELKEM ASA Research of Norway (R&D branch of a corporation with long, outstanding expertise in reactor design engineering), the Department of Chemical Engineering and the Department of Materials Science and Engineering at Carnegie Mellon University. The strategic goal of this partnership of academia with industry is a thorough technical and economic feasibility investigation addressing the proposed carbothermic production of aluminium; this ambitious objective clearly implies interdependent challenges, requiring theoretical and technical expertise at all levels, to ensure that (a) an interdisciplinary research team is assembled, (b) a realistic research program is outlined and executed, (c) clear objectives and success criteria are defined for all R&D stages, (d) regular progress evaluation provides future guidelines, and (e) usage of personnel, budget, and resources remains optimal.

The ongoing multidisciplinary and multifaceted research program encompasses a pleiad of concurrent necessary activities, including an extensive literature survey of journal papers as well as patents, a systematic thermophysical property compilation for chemical compounds, an evaluation of various previous computer modeling conclusions, a multilevel computational modeling program of the carbothermic reactor design conceived (using stage and total process models), laboratory-scale experiment/microscopy campaigns addressing the complex reaction mechanism and mass transfer at the microscale level, a multitude of large-scale pilot reactor experiment campaigns addressing many different complicated technical challenges, process operability, controllability and safety investigation, and detailed quantitative economic analysis.

The lasting alliance among ALCOA Inc., ELKEM ASA and Carnegie Mellon University forms an ambitious prototype for meaningful collaborations between industry and academia, clearly aimed at applied research and development of a novel process with a definite potential for construction and commercialization. The obvious challenge here and throughout the research program is the vital necessity to coordinate effort, disseminate information, set goals and evaluate progress so as to accelerate developments; the existence of several research units, groups and individuals with diverse backgrounds must be seriously considered, because it can pose challenges that are crucial to the effectiveness of the team and the success in the objective.
**Introduction**

Carbothermic reduction is a nontraditional alternative process for aluminium production, based on a high-temperature endothermic reduction reaction occurring between aluminium oxide and carbon. This process has an identified potential for drastic reduction of the fixed and operation costs associated with a greenfield investment. Furthermore, it is environmentally benign and in principle much more energy-efficient than conventional electrochemical methods, as the costly electrolytic reduction is effectively avoided using carbonaceous reactants via the direct chemical reduction pathway. This method is identified as a potential alternative to Hall-Héroult electrolytical reduction by several studies, but its complexity poses many remarkable technical obstacles for implementation. Therefore, if such an endeavor is to be pursued all the way to successful full-scale reactor construction and commercialization, it is in the best interest of the enterprise involved to undertake the R&D effort using all technical expertise available.

Interdisciplinary collaboration is a powerful way to draw from diverse resources, combining academic and industrial expertise. Thus, a pleiad of experts, researchers and modelers was convened to attend a workshop on carbothermic aluminium that was held in January 1999 in Pittsburgh under the auspices of ALCOA Inc. Following this event, a collaborative team was formed among Carnegie Mellon University, ALCOA Inc. and ELKEM ASA, its goal being to create an integrated process modeling environment for research, experimentation, development, design, control and economic analysis of a new carbothermic aluminium process R&D venture. The investigation of technical feasibility and economic viability of such an undertaking is the strategic objective of this partnership. A systematic approach to process design was strongly advocated by the academic experts in process modeling who joined the team. It is important to underline that systematic process development procedures are the definite standard in the petrochemical industry. Nevertheless, this common R&D practice is much less followed in the metallurgical industry, which is characterized by the vast complexity of unit operations and a predominantly iterative, experimental nature of novel process design efforts, both resulting in a lack of mathematical models that could facilitate R&D and systematic optimization endeavors.

The philosophy of the ongoing collaboration is to advance the carbothermic program efficiently by introducing Process Systems Engineering (PSE) and Computer-Aided Design (CAD) tools. Rather than resorting to the traditional empirical approach of an extensive, time- and capital-consuming experimentation program (which essentially probes the vastly multi-dimensional space of all design alternatives without any systematic engineering insight), the goal of the present collaborative effort is to combine strengths and interface researchers, knowledge, computer tools and data, so as to significantly accelerate the development of this novel process.

**Motivation and Historical Background of Carbothermic Processes**

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 aluminium from its compounds are generally far from economical [1]. The Hall-Héroult electrolytical aluminium oxide reduction process is recognized as the only technology for aluminium production that is technically feasible and economically profitable. Nonetheless, electrolysis of aluminium oxide is surely 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 concerns [3]. The direct chemical reduction pathway has been identified long ago as a less expensive and more benign process alternative which can improve total volumetric reactor productivity [4]. The importance of reducing the historically consistent sensitivity of the aluminium industry to energy prices is a major reason to invest in R&D of potentially profitable process alternatives.
The significance of this incentive is clearly illustrated by the impact of a recent energy crisis on aluminium production in Figure 1(a): monthly averages of U.S. primary aluminium production plummeted by more than 28% between the years 2000 and 2001, as a result of the major energy crisis that struck the United States in early 2001, inducing major disruptions in production [5]. Aluminium industries have thus championed R&D of new carbothermic processes, particularly during energy crisis periods; this explains why the investigation begun several decades ago [6]. The patent literature is the primary source of historical details regarding the evolution of several carbothermic aluminium reactor concepts. The comprehensive patent literature overview by Motzfeldt et al. [4] provides a chronological summary of main technical innovations and points out many obstacles posed by the nature of patents which also hinder remarkably R&D efforts (patent language and documentation usually pose challenges to creating a sound design basis).

A significant interest in the carbothermic production of aluminium occurred at the end of the 1950s and the beginning of the 1960s: three major aluminium producers (ALCAN, Pechiney, Reynolds) undertook large-scale testing projects to investigate the potential of carbothermic chemistry, while ALCOA had also started its own smaller-scale testing a few years earlier [4].

ALCAN developed a carbothermic process suitable for producing Al: the reactions were staged in separate chambers and electric heating was achieved through the melt. Publications did not appear in the literature, and the process was abandoned due to construction material problems. Pechiney entered the quest for carbothermic aluminium processes and built a plant at Nogueres, France (1960) that was operated until 1967. Technical details were only published decades later by Crussard, and Legrand and Leroy (see Ref. in [4]). The process used is assumed [4] to have occurred in three stages: first, bauxite was partly reduced with C in an electric furnace; then, the produced Al₂O₃ was reduced with C to tap impure Al; finally, C impurities were removed from Al as Al₄C₃, via cooling. Pechiney patented and described a variety of proposed reactors. Reynolds Metals Co. was among the pioneers of research efforts, with an extensive experimental and modeling program undertaken at an Alabama laboratory facility (1971–1990). The detailed investigation resulted in several notable technical reports and publications [7, 8, 9]; it also identified serious technical challenges (showstoppers) for a viable and profitable process. Technical obstacles were anticipated to hinder the laboratory-scale to industrial-scale transition: vaporization of gaseous Al species produced by slag superheating at high temperatures made it clear that reaction staging (and ensuing Al(g) recovery and recycling) are elemental to viability.

Kusik et al. of Arthur D. Little, Inc. later published the significant conclusions of a thorough technoeconomic assessment study commissioned by the U.S. Department of Energy [10]. 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” encouraged further pursuit of this ambitious endeavor. ALCOA Inc. and ELKEM ASA thus spearheaded R&D in the field, and the collaboration led to a multidisciplinary R&D effort [11].

Figure 1: (a) Energy crisis vs. aluminium production, (b) academic R&D funding sources [12].
Carbothermic Reactor Engineering

Carbothermic reduction is the only non-electrochemical process that has been proposed and tested for aluminium production [2], and can be carried out as two-stage reaction process. Carbon has been proposed as a reducing agent, since a plethora of experimental studies have documented that even its relatively low-purity forms can effectively advance reduction [8, 9]. Carbothermic reduction of alumina to aluminium is extensively investigated: the literature survey reveals numerous studies conducted at the laboratory or pilot-plant scale [4, 7-9, 13-15]. The high reaction temperatures entail the use of electric energy for heating, but a carbothermic process is more energy-efficient and exhibits notably high theoretical volumetric productivity. Thus, considerable industrial reactor sizes and significant economies of scale can be achieved. A wide variety of industrial scale reactor designs have been proposed in the literature [4], but we focus on ARP [16], the conceptual carbothermic reactor design of Johansen and Aune [17]. Although the theoretical foundation of process chemistry is sound, implementation details set the stage for feasibility challenges, which are due to the technical difficulties associated with handling multiphase molten slags and to the extremely high temperatures (> 2000 °C) required. A schematic representation of the proposed reactor is provided in Figure 2: a clear observation is the effort to take advantage of the chemistry by separating phenomena in four distinct stages.

1. The first stage of the process is a pre-reduction smelting zone: C and Al₂O₃ pellets are continuously fed to an open arc electric smelter, melt and react to form a viscous binary slag. The reaction of Al₂O₃ with a C excess to form the Al₄C₃-rich slag is written as (T > 1900 °C):

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

2. The second stage is the high-temperature reduction zone: the first-stage molten slag flows slowly into a second multi-electrode submerged arc reactor, and it is further heated to a higher temperature, avoiding local surface superheating. Liquid Al and CO gas are rapidly generated; Al₄C₃ injection assists in shifting the equilibrium towards Al and in avoiding carbon depletion. The decomposition of the Al₄C₃-rich slag towards the Al-rich phase is written as (T > 2000 °C):

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

3. The third stage consists of a Vapor Recovery Reactor (VRR), where Al and Al₂O vapors (both inevitably produced in any high-temperature reactor due to localized superheating) react with C to form Al₄C₃. Unless Al(g) species are recovered countercurrent to the incoming feed, Al(l) loss shifts the chemical equilibrium with a catastrophic impact on process economics [18].

4. The fourth stage of the process is the purification zone: the liquid Al alloy (of lower density) produced in the second stage floats as a top layer and flows through an overflow weir to a tank, where dissolved C and entrained Al₄C₃ particles are then removed to recover pure aluminium.

![Figure 2: Schematic reactor and model representation for the ARP carbothermic process [19].](image)
The D.O.E. Carbothermic Aluminium Research Program

The U.S. federal government remains the primary funding source for academic research, but at the firm decrease rate of Figure 1(b) [12]; industrial funding for academic R&D work has however increased steadily, encouraging institutions to benefit from interesting opportunities. Our collaboration resulted from a joint R&D proposal for an Advanced Reactor Process (ARP). ARP is a multi-step high-temperature reaction technology [11] producing aluminium by direct reduction of alumina with carbon. R&D of a continuous multi-zone furnace able of producing the high temperatures required and recovering molten and gas phases is critical. This is Phase I of a multi-phase effort to develop ARP, based on expertise on advanced reactor technology and an effort for better understanding of underlying reactions and thermodynamics. The goal of the carbothermic R&D project is thus to demonstrate the technical, economic and environmental viability of aluminium carbothermic technology by using advanced high temperature (2,000 °C) bench- and laboratory-scale reactors. The project team faces a need to address technical hurdles which have repeatedly prevented full development and implementation of this technology in the past. The hurdles include the high temperatures required and their effect on: (a) the recovery of aluminium, (b) the selection of construction materials for reaction zones, and (c) the reduction of Al₄C₃ impurity in the final aluminium product. The most promising process options will be selected for further investigation in the ensuing large-scale reactor testing campaign (Phase II).

Although the carbothermic process involves the generation of carbon-based greenhouse gases (GHG), the total GHG reduction from power plant to metal anticipated from implementing this technology will be substantial due to: (a) the significantly reduced power consumption, and (b) the elimination of perfluorocarbon emissions as well as carbon anode baking furnace emissions. The estimated capital investment required for ARP will be about 50% less than that for conventional Hall-Héroult cell technology; the power consumption is ca. 8.5 kWh/kg, inducing an estimated 25% manufacturing cost reduction; the required labor will be reduced as well [11].

Carnegie Mellon University: Research Objectives

The strategic goal of this collaboration from the perspective of the Department of Chemical Engineering at Carnegie Mellon is to offer an integrated computer simulation framework for carbothermic reactor design that can radically reduce cost and expedite process development. Numerous process components have been implemented at laboratory or pilot-plant scale, but a carbothermic aluminium reactor has not been tested yet and a lot of uncertainties still remain. Four complementary systems approach goals are pursued to advance with integrated modeling:

1. The development of a variety of customized process models (steady-state, dynamic, algebraic/DAE, distributed/PDE models) addressing each of the process stages as well as the total process; also, the development of optimization (MINLP) models addressing economic viability with suitable objective functions. Model building and development issues are many, but the fundamental questions are always two: “What information do we want the model to provide? Why and how is this information necessary to allow advances with the design effort?”.

2. The efficient integration of commercial process simulation tools, thermophysical property databases and experimental data: this is in fact a crucial task in avoiding oversimplifications. An illustrative example related to this goal is the need to include computationally intensive free energy minimization calculations in the core of macroscopic process optimization models [19]: the thermodynamic equilibrium assumption is realistic but entails software interfacing problems due to formulation complexities. This challenge is also a strong motivation towards pursuing multiscale modeling, a new powerful systems concept that can address complex problems [20].

3. The identification of the most sensitive variables whose study must be actively pursued for experiment and pilot plant design, to avoid exhaustive search means and economize on R&D cost. Sensitivity analyses can quantitatively evaluate the importance of uncertain process
parameters and provide sound guidelines for efficient experiment design, economizing on expenditure. Furthermore, they are used to evaluate the relative importance of experiments and point the regions of physical space that should be probed to generate knowledge about physical parameters. The multi-dimensional space of all design alternatives is not subject to exhaustive search strategies if CAD tools are used; engineering insight can emerge in a systematic way. PDE modeling and CFD simulation can facilitate experiments and preliminary design [21-22].

4. The creation of a novel computational environment for integration of archived and newly acquired knowledge from ongoing work, modeling, experiment design and evaluation, process design, optimization, and detailed economic analysis. Considerable knowledge about physical and economic aspects of carbothermic processes is compiled but remains distributed, and it is remarkable that this problem hinders design efforts, as it poses challenges to creating a sound and accessible design basis. This latter goal is indispensable to the optimal use of resources, and a reality we have repeatedly experienced in the carbothermic aluminium research program. Thermodynamics, kinetics, thermophysical material properties, computational fluid dynamics, large-scale numerical solution and optimization strategies, economic evaluation methods and costimation databases constitute facets of a unified environment for rapid evaluation of different process alternatives. The technical and economic feasibility of conceptual process configurations is to be repeatedly evaluated against many performance parameters over time, as measurements become available. The alternatives demonstrating the greatest promise for viability, operability, controllability, safety and environmental advantages form a basis for model refinement, extensive pilot plant testing and construction engineering. The iterative nature of this procedure demonstrates the extreme value of the existence of a unified modeling and evaluation environment and illustrates IT integration needs of modern enterprises [23-24].

Undertaking the modeling project is part of a larger effort geared towards process design, optimization and distributed simulation, and involving several faculty members and doctoral students in the Department of Chemical Engineering at Carnegie Mellon and the Center for Advanced Process Decision-Making (CAPD). Lasting expertise in large-scale process design and optimization is taken advantage of, in order to pursue theoretical directions: we explore how advanced information storage/retrieval systems and in-house numerical methods of Carnegie Mellon University can effectively support real-world process design ventures by modifying and updating models of many process configurations. Doing so in a seamless fashion (as better and more accurate data becomes available from experimentation) is no less important: advanced computer models are meaningful only when acceptably linked to experimental tests. Application of CAD and systems engineering tools to the carbothermic project thus allow us to:

- Develop stage process models independently and study phenomena in a distributed fashion.
- Pursue gradual model elaboration and tailor model development to most crucial aspects.
- Simulate the static and dynamic operation of process stages and the interconnected system.
- Pinpoint experiments and parameters that enhance understanding of underlying phenomena.
- Assist the pilot-plant experiment design by performing series of related “what-if” analyses.
- Provide reliable balance calculations to permit accurate cost estimates at every design stage.
- Develop a complete design record and provide a sound design basis for future investments.
- Develop case studies in order to guide ALCOA and ELKEM in broader CAD collaboration.
- Create the unified computational modeling environment for design and simulation of ARP.

A significant and expensive development effort is clearly needed before concluding anything about commercial process viability. A comprehensive overview of the organization and growth of research activities and the expertise generated during the process is presented in the following section, and relevant interdependence challenges illustrate collaboration complexity.
Carbothermic Aluminium R&D Program: Activities and Contributions

The carbothermic aluminium research program encompasses a wide spectrum of R&D activities which are equally important to the development of a feasible as well as viable reactor design. Thus, while keeping in mind the importance of interfacing researchers in order to communicate results and conclusions, it is crucial to assign tasks, delegate activities and allocate resources to make the best possible use of expertise. Consequently, numerous responsibilities have been undertaken and a number of publications (about problems, ideas, challenges and achievements) have emerged. The present partnership, in itself, is thus a model for authoring original papers without compromising on the confidentiality constraints on proprietary technical information.

Thermophysical properties (density, viscosity, thermal and electric conductivity, diffusivity) are fundamental thermodynamic characteristics of phases that have a dominant effect on reactor design. Thorough understanding of physical properties is necessary to overcome barriers that halted progress in the past [7-10], and property measurement and/or estimation is therefore critical to ARP success. A comprehensive review [21] of references on molten slags in general [25-26] and the Al₂O₃-Al₄C₃ system in particular [27-29] led to a compilation of correlations [21] that we have already incorporated successfully in our first-principle CFD simulations [22].

The exceptional importance of molten slag viscosity should be discussed in particular, because this transport property affects simultaneously a variety of physical phenomena in the reactor, namely: (a) the macroscopic slag flow nature (Re number), (b) the melting and solidification of slag against reactor walls, (c) the phase distribution in the bulk of the carbothermic reactor, and because of it (d) the heat transfer rate across all interfaces. The temperature dependence of slag viscosities is exponential (quasi-Arrhenius) in most of the phenomenological models [25-26], rendering estimation uncertainties catastrophic. Developing a precise viscosity prediction model (ideally as a function of slag composition and temperature) is essential, but requires explicit knowledge of all ionic species and of the molten slag ionic structure, as pointed out in a study by the Department of Materials Science and Engineering at Carnegie Mellon University [30].

A systematic finite element CFD modeling and simulation study of the core (second stage) of the proposed carbothermic reactor has also been pursued consistently during the years [20-22] in order to develop reliable CFD models and methods that will allow us to conduct sensitivity analyses for identifying key design variables. The effectiveness and reliability of the new thermophysical property correlations is confirmed by the accuracy achieved after incorporating the aforementioned property models in our CFD calculations [22]. A very important remark regarding the reliability of CFD models is their capability of reliably reproducing operating temperatures for the second reactor stage without any experimental input [22]. The use of PDE modeling tools and such detailed computations is subject to parametric and modeling assumption uncertainties; however, early success is encouraging to advance with our goals. The integrated electric charge, heat and momentum balances are solved by a FEMLAB® model [31].

A powerful modeling idea we have also proposed and pursue is the creation of an effective multiscale modeling hierarchy [20]: the idea there is to expand the standard mass and heat balance calculations of macroscopic nature [19] one step further, toward enhancing accuracy via microscopic PDE calculations [21-22]. The goal is to solve the full steady state PDE problem (electric charge, heat, momentum and molar species balances) for the respective state variables [potential (V), temperature (T), velocity (Uₓ, Uᵧ, Uₗ), pressure (P), concentrations (Cᵢ)] and use the resulting distributions as design guidelines (such real-time CFD computations are not tolerable because of extreme CPU requirements). To perfect the CFD model reliability and allow for meaningful component concentration predictions, it will be essential to use sophisticated thermodynamic equilibrium calculations software performing Gibbs free energy minimization (FACTSage®, [32]), hence in fact incorporating chemistry to an abstract CFD model. The latter implies formal representation complications, generating clear and inevitable software interfacing challenges. Even when this acute problem has been successfully addressed,
experimental validation of CFD and design model simulations is vital to ensure their reliability and prediction potential. Once the latter are established, models allow for predictive process performance calculations at various operating conditions, resulting in insight for improved design, operation and control. The development of a total process model is a desirable option.

Elaborate experimental reactor development and investigation is obviously required, to support and perfect computer simulation models, and collect valuable measurement data under actual operation. The importance of the latter is paramount for high-temperature processes: operation cannot be replicated at the laboratory scale. This is especially true for reaction mechanisms and kinetics; therefore, a laboratory-scale VRR was used to examine and identify the controlling step during Al(θ) recovery [33]; results in Figure 4 are in agreement with model predictions [34]. An extensive reactor testing experimental program has also been undertaken by ELKEM ASA Research at their R&D facility in Kristiansand, Norway, to achieve and study reactor operation. The goal of the experimental pre-pilot plant test campaign series has been to develop a continuous electric arc furnace process by individually constructing, testing, troubleshooting and evolving the four reactor compartments (the ARP stages) in a concurrent, modular fashion. Modular R&D and testing is followed by an integration phase to link stages in a single process. Numerous challenges are recognized and documented as critical to efficient operation [35-36].

Figure 3: Elect. potential ($V$), intensity ($E$), temperature ($T$) and velocity ($U$) distributions [22].

![Figure 3: Elect. potential ($V$), intensity ($E$), temperature ($T$) and velocity ($U$) distributions [22].](image)

Figure 4: Dynamic process model of a Vapor Recovery Reactor and representative results [34].

![Figure 4: Dynamic process model of a Vapor Recovery Reactor and representative results [34].](image)
Conclusions

The international, interdisciplinary nature of this collaboration between industry and academia for new process development creates a variety of technical and communication challenges, but also explores unchartered areas of process design for high-temperature metallurgical processes. The systems approach draws from a conceptual design basis and presents a paradigm for coordinating R&D and modeling efforts, to tackle pressing challenges with reliable CAD tools. A unified computational modeling environment is the ultimate goal for systematizing design.

Future Goals: Economic Evaluation and Commercialization

The touchstone of economic viability and financial profitability of a venture is a thorough economic evaluation, whose reliability depends on the accuracy of engineering calculations. The latter pertain to mass and heat balances and permit capital and operating cost estimations. Quantitative assessment expertise accumulated at ALCOA Inc. is a guarantee toward reliable commercial-scale cost estimations for the ARP process. Nevertheless, the challenge to create a suitable computational environment for multiple “what-if” scenario analyses remains, because of the vast number of uncertainty sources illustrated in Figure 5. The relative importance and impact of such uncertainty factors can be quantitatively analyzed using tornado diagrams [24]. Prospective commercialization criteria have been defined [12]: (a) a 50% capital cost reduction over the Hall-Héroult process, (b) a 25% production cost reduction, and (c) an environmentally acceptable system. A commercialization plan for efficient marketing has also been developed.

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