A MULTISCALE MODEL FOR CONCEPTUAL DESIGN AND SIMULATION OF A CARBOTHERMIC REDUCTION PROCESS FOR ALUMINIUM PRODUCTION

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Abstract

Multiscale modeling is a tool aimed at combining mathematical descriptions of different process scales into a single, properly tailored scale integration hierarchy allowing for reliable modeling and simulation. This concept has already been successfully applied in deriving accurate state variable distributions for processes that exhibit full spatiotemporal variation and thus do not allow for the standard simplifications. This paper discusses the fundamental considerations required for the development of a multiscale model and presents the main implementation challenges for a conceptual high-temperature multiphase reactor. The objective is to simultaneously solve the electric charge, heat, momentum, mass and molar balances for the carbothermic reduction of alumina, a complex electrochemical process for aluminium production. This multiscale model relies on a decomposition of the PDE (mass, heat and momentum) balances into two levels: the first modeling level consists of a series of CSTR reactors resembling a conceptual reactor, and the second modeling level uses a Computational Fluid Dynamics model for each of those sections. The main contribution of our study is that it proposes a framework for solving complex process problems of the metallurgical industry, where the spatially distributed state variables are of particular importance.

Keywords

Multiscale process modeling, computational fluid dynamics (CFD), carbothermic aluminium production.

Introduction

Multiscale hierarchies of mathematical descriptions which all address different phenomena present in a single process have emerged as powerful tools for efficient simulations in the conceptual design of challenging chemical processes. The importance of multiscale modeling is particularly exemplified in novel and complex multiphase applications, which cannot admit the standard modeling approximations. Atmospheric pollution modeling is one of the first fields that benefited from multiscale modeling ideas (Odman and Russell, 1991; Kumar et al., 1994; Kumar et al., 1997). Advanced semiconductors and chemical vapor deposition for new materials are also active fields in which multiscale modeling has been fruitful (Srolovitz et al., 1997; Rodgers and Jensen, 1998; Gungor et al., 1999; Maroudas, 2000).

Multiscale modeling for process systems engineering relies on reactor zone models (Bermingham et al., 1998) coupled with spatially detailed PDE descriptions, to obtain detailed solutions for flow fields and/or size distributions. Recent publications include work on diverse applications which are all characterized by their distributed nature and apparent complexity – both necessitating spatial accuracy. A novel multistage crystallizer (Urban and Liberis, 1999), a gas/liquid column reactor (Bauer and Eigenberger, 1999) and a semi-batch fermentation reactor (Bezzo et al., 2000) are illustrative processes that were successfully explored. The latest extensive review (Pantelides, 2000) provides the fundamental reasoning for multiscale process modeling and discusses problems and challenges in this broad area.

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Atmospheric pollution multiscale modeling has been historically a fruitful approach, as it takes advantage of the explicit knowledge of all atmospheric kinetic mechanisms, combined with large-scale finite element flow models, so as to study air quality as a function of pollutant emissions and derive predictions which can be validated via satellite observation methods reviewed by Singh and Jacob (2000). Advanced materials modeling has been equally successful, as it employs molecular simulation methods combined with computational continuum mechanics (Maroudas, 2000) to derive detailed characterizations of various microstructures and allow for quantitative prediction of material behavior. The latter are validated by many experimental techniques.

Multiscale Modeling for Chemical Process Design

Process systems design and simulation is a realm in which multiscale modeling faces more pressing challenges: the application of multiscale modeling concepts to process design is a relatively novel venture, as physical phenomena and numerical problems hinder efficient implementations. In contradistinction to atmospheric and materials problems, modeling for process systems simulation has to focus on phenomena at the intermediate length and time scales. Various three-dimensional incompressible reactive flows define a spectrum of challenges in atmospheric modeling, while molecular structure and performance in applications are central issues to heterogeneous materials modeling.

Because the interplay between transport phenomena, (e.g. convection occurring within various unit operations) and rate phenomena (e.g. multiphase chemical reactions) is tightest at those very intermediate length and time scales, implementation of multiscale process models is intricate. Thus, the challenges already identified (Pantelides, 2000) are to first draw rules for efficient phenomena partitioning among the computational modules (scale embodiments), and then address numerical stability and efficiency issues (a successive substitution procedure is not always reliable).

Carbothermic Production of Aluminium

Carbothermic reduction of alumina to aluminium is a high-yield, economically favorable and environmentally benign process with a potential for industrial application. Reynolds Metals Co. identified it as an alternative to the Hall-Héroult electrochemical reduction (Motzfeldt, 1989), as ionization is effectively eliminated by carbon reduction. The conceptual multistage electrothermic reactor jointly proposed by ALCOA and ELKEM (Johansen et al., 2000) is an attractive idea for achieving reactor scaleups that will unleash the significant volumetric production advantage. Nonetheless, the complexity of carbothermic reduction and the extremely high temperatures required (T > 2000 °C) pose remarkable technical challenges for implementation. The distributed nature of the design is the most interesting, as the electrothermic heating necessary for endothermic reaction is achieved by independent AC electrode pairs. Concurrent multiphase phenomena induced by the reaction (generation of CO gas bubbles and Al liquid droplets) also introduce modeling complications and control challenges.

The conceptual carbothermic reactor considered in this work is illustrated in Figure 1 (Johansen et al., 2000). This carbothermic reduction reactor features four stages; relevant details regarding its development and operation have been published elsewhere (Gerogiorgis et al., 2001). The present study focuses entirely on the second stage and inevitably employs a set of simplifying assumptions so as to facilitate the development of a multiscale design model. Thus, we consider perfect mixing at the first (CSTR) level, include the Joule heating effect in mass and heat balances and use simplifying approximations for chemical reactions. A second (CFD) level solves the Navier-Stokes problem for the (assumed) three-dimensional laminar slag flow field (with no transition to turbulence because of gas formation). An assumption of instant thermodynamic equilibrium can be invoked to model high-temperature phase coexistence.

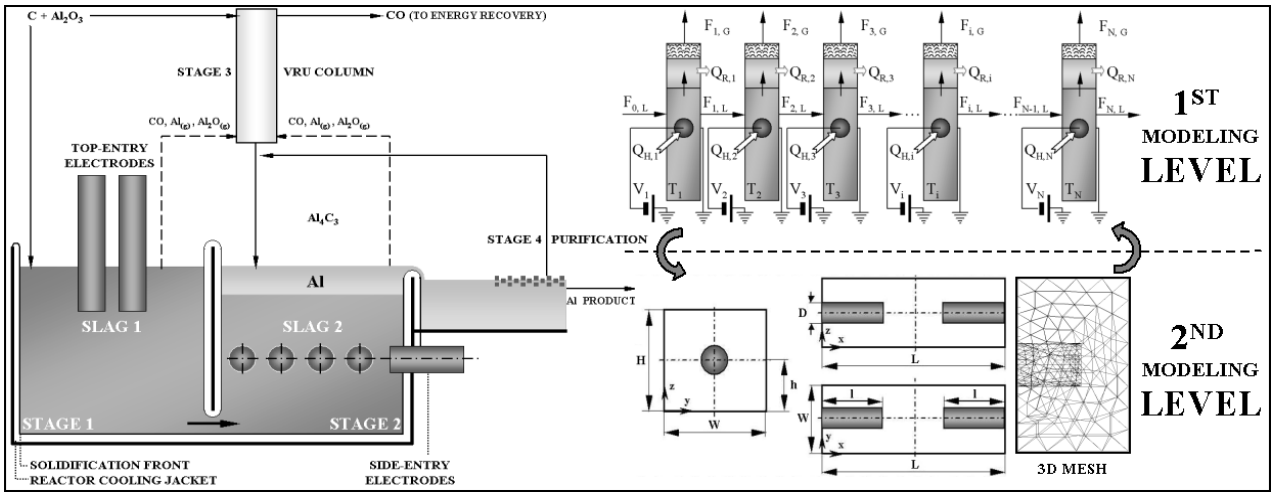

Figure 1: A conceptual carbothermic reactor (Johansen et al., 2000) and the corresponding multiscale model.

Multiscale Model

The multiscale process model presented here has been developed for a conceptual carbothermic reactor (Figure 1) and is constructed at two complementary interacting levels. The first level (a series of N steady state CSTR reactors) represents the configuration of the carbothermic reactor, solely focusing on its second (aluminium-producing) stage. The principal horizontal flow axis is used for discretization under the fundamental assumption of perfect mixing for each of those vertical sections; all are modeled as CSTRs. The second level (a Computational Fluid Dynamics model) focuses on each of the N reactor sections, considering the steady state three-dimensional partial differential equations (PDEs) that describe charge, heat and momentum balances. The incompressible Navier-Stokes flow problem is solved considering Boussinesq (buoyancy) momentum generation as a result of the temperature-variant liquid slag density. Constant thermophysical slag properties are assumed, and models thereof are compiled by Gerogiorgis et al. (2001). A temperature-dependent electrical conductivity model is used to illustrate the strong coupling between the potential gradient and the Joule generation term in the heat balance. Slag melting and solidification at the cooled reactor walls as well as population balances (CO bubbles, Al droplets) are not included here, because of parametric uncertainty, but can in principle be accommodated within this model. The finite element method is used (Hanke, 2000) to derive a solution to the coupled PDE problem. The model is implemented on an unstructured tetrahedral discretization of the domain (Figure 1), using quadratic basis functions.

The goal is to solve the full steady state PDE problem for the respective variables of the balances [potential (V), temperature (T), velocity (U_x ,U_y ,U_z), pressure (P)] and use the resulting profiles interactively with first level results, to form a loop for iterative calculation of spatial distributions. Thus, first level results can serve as boundary conditions that are reliable approximate inputs for the second model. Suitable integrations of second level model results in turn serve as first level inputs when convergence is achieved. For example, boundary integration of velocity yields flows and the respective one for temperature yields heat flows.

This strategy is similar to that of Urban and Liberis (2000) in that our first level resembles their "zone model" – generally defined arbitrarily, based on reactor symmetry, technical experience and/or experimental observations. Coupling a zone model with a CFD model has also been used for efficient dynamic simulation (Bezzo et al., 2000); however, the study of complex processes often requires to probe more spatial variables than temperature and velocity. Accordingly, the modeling challenges here are many more:

- carbothermic reduction chemistry is uniquely complicated
- electrothermic heating (the Joule effect) induces a strong nonlinear coupling between the charge and heat balances
- slag viscosity is quite carbon- and temperature-dependent
- finally, the extreme temperatures yield physical property uncertainties by rendering measurements hard and costly.

Model Implementation and Results

The first modeling level is implemented in MATLAB® for solving the steady state nonlinear algebraic system. Selecting the independent electrode pair voltages as inputs (as voltage can serve as a convenient manipulated variable) and assuming field intensity is constant within each section (in addition to temperature and all species' concentrations) can yield the respective liquid/gas piecewise flow profiles for each assumed CSTR along the horizontal reactor axis. The accuracy of the results obtained strongly depends on the thermodynamic phase diagram (Motzfeldt et al., 1989) and the liquidus curves therein (Gerogiorgis et al., 2001), but also on the simplified kinetic models derived from the Arrhenius plots published by Motzfeldt for the gas species. An instant thermodynamic equilibrium assumption is more accurate in principle, but a third (free energy minimization) level would be necessary to achieve closure in that case.

The second modeling level (FEMLAB® v. 2.2) uses half of the CSTR section of the multielectrode furnace, employing the symmetry across the horizontal reactor axis and the assumption of a homogeneous Al_2O_3-Al_4C_3 slag, for a domain comprising a C electrode submerged therein.

The computational challenge in this multiscale model is to exchange intermediate results in a seamless fashion. The single platform shared by MATLAB® and FEMLAB® thus allows efficiency in steady state process simulations.

Figures 2 and 3 depict the state variable distributions obtained for the middle of the reactor (XZ and YZ sections are taken at the electrode midplane and tip, respectively).

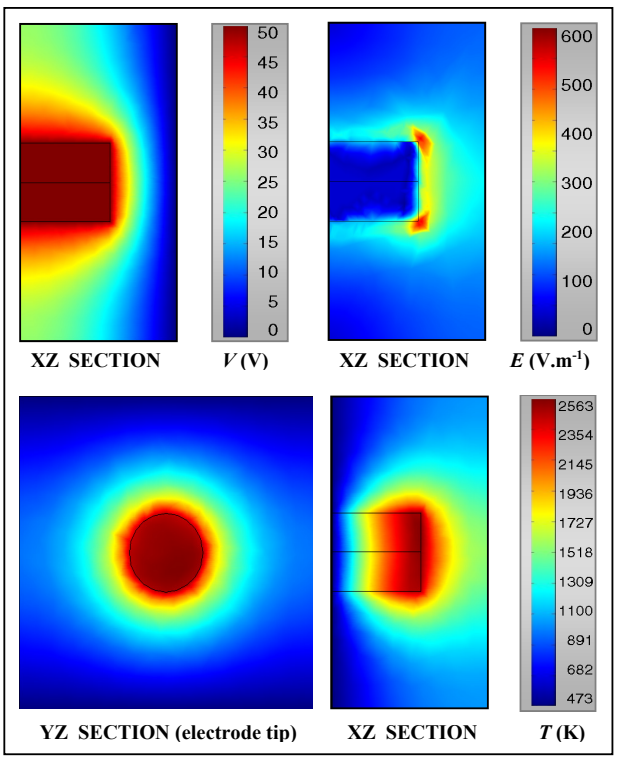

Figure 2: Multiscale model results (Joule heating).

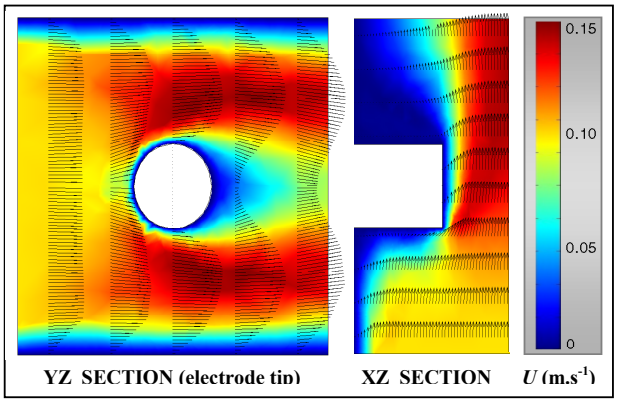

Figure 3: Multiscale model results (slag flow field).

Conclusions

A two-level steady state multiscale process model has been constructed for a conceptual carbothermic reactor by combining a steady state CSTR series model (first level) with a finite element PDE balance model (second level). The charge, heat, momentum, mass and molar balances can be solved within this framework by a scale integration, by using results of one model as input updates to the other. The representative steady state profiles from the first level as well as the spatial distributions from the second level show that the model provides results of adequate accuracy. The potential profile from the Laplace Equation ($\nabla^2 V = 0$) can be solved independently; gradient accuracy is crucial though, as the electric field intensity ($E = \nabla V$) is coupled with the heat balance Joule production term (Figure 2). The temperature profile obtained is presented in Figure 2. The velocity profile barely reveals a vertical recirculation as hotter slag rises (Figure 3), but the uncertainty related to thermal expansion coefficient (α) value is important; thus, quantifying the Boussinesq effect is still an open problem. Model predictions are in good agreement with qualitative observations so far; however, experimental validation is still very difficult in the high-temperature complex reactor.

Although significant approximating assumptions have been made to reduce its size, this model can in principle accommodate augmented multiphase process descriptions, if models and parameters are available for the phenomena. The distributions can be used for comparative evaluation of candidate reactor designs; a quantitative evaluation of the model's computational efficiency is also under way.

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References

Bauer, M., Eigenberger, G. (1999). A Concept for Multi-Scale Modeling of Bubble Columns and Loop Reactors. *Chem. Eng. Sci.*, *54*, 109.

Bermingham, S. K., Kramer, H. J. M., Van Rosmalen, G.M. (1998). Towards On-Scale Crystalliser Design Using Compartmental Models. *Comput. Chem. Eng.*, *22*, 355.

Bezzo, F., Macchietto, S., Pantelides, C. C. (2000). A General Framework for the Integration of Computational Fluid Dynamics and Process Simulation. *Comput. Chem. Eng.*, *24*, 653.

Gerogiorgis, D. I., Ydstie, B. E., Seetharaman, S. S. (2001). A Steady State Electrothermic Simulation Analysis of A Carbothermic Reduction Reactor for the Production of Aluminium. *In Proceedings of the Meeting on Computational Modeling of Materials, Minerals and Metals Processing*. TMS Annual Meeting, Seattle, 273.

Gungor, M. R., Gray, L. J., Zhou, S. J., Maroudas, D. (1999). Modeling of Failure in Metallic Thin Films Induced by Stress and Electromigration: A Multiscale Computational Analysis. *Mater. Res. Soc. Symp. Proc.*, *538*, 263.

Hanke, M. (2000). Towards a New Implementation of a Nonlinear Solver for FEMLAB®, Technical Report, Royal Institute of Technology, Stockholm, Sweden.

Johansen, K., Aune, J. A., Bruno, M., Schei, A. (2000). Carbothermic Aluminum – ALCOA and ELKEM's New Approach Based on Reactor Technology to Meet Process Requirements. *In Proceedings of the 6th International Conference on Molten Slags, Fluxes and Salts*. Stockholm, Sweden, paper #192 (CD-ROM).

Kumar, N., Odman, M. T., Russell, A. G. (1994). Multiscale Air Quality Modeling: Application to Southern California. *J. Geophys. Res.*, *99(D3)*, 5385.

Kumar, N., Segall, E., Steenkiste, P., Russell, A. G. (1997). Parallel and Distributed Application of an Urban-to-Regional Multiscale Model. *Comput. Chem. Eng.*, *21*, 399.

Motzfeldt, K., Kvande, H., Schei, A., Grjotheim, K. (1989). Carbothermal Production of Aluminium. *Aluminium Verlag*. Dusseldorf, Germany.

Maroudas, D. (2000). Multiscale Modeling of Hard Materials: Challenges and Opportunities for Chemical Engineering, *AIChE Journal*, *46(5)*, 878.

Odman, M. T., Russell, A. G. (1991). Multiscale Modeling of Pollutant Transport and Chemistry. *J. Geophys. Res.*, *96(D4)*, 7363.

Pantelides, C.C. (2000). New Challenges and Opportunities for Process Modelling. *In Proceedings of the European Symposium on Computer Aided Process Engineering – 11 (ESCAPE–11)*. Kolding, Denmark, 15.

Rodgers, S. T., Jensen, K. F. (1998). Multiscale Modeling of Chemical Vapor Deposition. *J. Appl. Phys.*, *83(1)*, 524.

Singh, H.B., Jacob, D.J. (2000). Future directions: satellite observations of tropospheric chemistry. *Atmos. Environ.*, *34*, 4399.

Srolovitz, D. J., Dandy, D. S., Butler, J. E., Battaile, C. C., Paritosh, F. (1997). The Integrated Multiscale Modeling of Diamond Vapor Chemical Vapor Deposition. *Journal of Metals*, *49*, 42.

Urban, Z., Liberis, L. (1999). Hybrid gPROMS-CFD modelling of an industrial scale crystalliser with rigorous crystal nucleation and growth kinetics and a full population balance. *Paper presented at the Chemputers 1999 Conference.* Dusseldorf, Germany.