Modeling of the Silicon Process as a DAE System

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ABSTRACT

A new approach towards modeling the silicon production process is investigated. A nonlinear dynamic model extends the Unidimensional Dynamic Model proposed by Halvorsen. The equations presented are a set of nonlinear partial differential equation representing mass and energy balances for the overall furnace. Algebraic constraint equations further complement the differential equation by representing kinetic rates, physical property definitions, thermodynamic expression, *etc*. An essential difference with our approach is the method of discretization, formulation and solution of the differential system. Method of lines (MOL) utilizing finite differences is employed to discretize the spatial part of the partial differential equations. Including the boundary conditions as algebraic constraints further alters the resulting initial value ordinary differential equation system. This formulation generates an efficient differential and algebraic equation (DAE) system. Mainly advantageous for its robust nature in conveying the physical meaning of the realistic process, a specially designed solver package aimed at solving DAE system is employed. We briefly describe the state of the art public domain solver, DASPK 3.0, which is a prominent tool with excellent numerical convergence and stability properties suited for solution of this problem. Simulation case studies are included and stress a comparison between finite difference approximations utilized.

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1 INTRODUCTION

The commercial silicon process has its roots dating back to the late nineteenth century. Since then, the process has undergone numerous developments in operational efficiency, size, electrode configuration, process control, etc., but the main principles to this day remain unchanged. Industrial experience shows that the silicon process is sensitive to fluctuations in the composition and quality of the feedstock. These disturbances have a crucial impact on the product (silicon) yield, purity and overall process stability. Current metallurgical control systems cannot cope with these disturbances alone, and experienced operator intervention is necessary to maintain process stability and silicon yields at acceptable levels. One of the main difficulties lies in the inability to measure specific physical quantities, which need to be taken into account in the process control operation.

With recent advances in modern technology, an increased interest has spurred in developing a detailed knowledge about the industrial silicon production process. Mathematical modeling efforts of varying complexity are intended to generate results that predict the furnace behavior based on any set of disturbances applied to the furnace.

Control of electrical load, current and resistance in the silicon furnace, has been well developed and numerous reports exist in this area. Metallurgical control for adjusting feed materials in order to maintain a high silicon metal yield and an internal specie concentration balance is not presently as sophisticated and will be the subject of future studies.

The main goal of the current study is to develop a robust nonlinear dynamic model utilizing recent advances in process systems, applied mathematics and computer science. Specifically, the model equations are discretized via method of lines (MOL) approach, the resulting set is formulated as a differential algebraic equation (DAE) system, which in turn is solved by a specially adapted solver, DASPK. The nonlinear model developed will be the basis for developing metallurgical control strategy for the silicon furnaces based on a real time optimization (RTO) framework. In RTO, the nonlinear dynamic model is responsible for predicting accurate furnace behavior and the optimal control moves are calculated using on-line optimization subject to an appropriate (economic) objective [11].

Models of varying descriptive complexity have been proposed for the metallurgical/chemical production process of (ferro)silicon metal. The model proposed by [15], considered a two-zone representation of the furnace, establishing the importance of the carbon reactivity and low SiO content in the lower (higher temperature) zone. A qualitative model developed by Müller *et al.* [12] explains the high reaction rate between the condensed materials, SiO_2 and SiC, by evaporation of Si. The Si evaporates in the arc and the vapor diffuses to the SiO_2 particles where it reacts to produce SiO, which reacts further with SiC to make Si.

Budde *et al.* [5] have constructed a model where the furnace is composed of interconnected system of reaction zones. Balance equations for the charge, energy, momentum and mass were incorporated and numerically solved. Despite providing graphical results for spatial distribution of loads and temperature, the authors were unclear as to their treatment of chemical kinetics and the associated parameters. Nevertheless, the approach reveals a study between chemical reactions and electrical current in the system.

A model developed at KemaNord [15] in Sweden, describes a furnace as a set of vertically stacked segments combining equilibrium calculations with heat and mass balances. A computer program designed to calculate a solution for each segment, minimizes the free energy. Chemical kinetics are treated partly by selecting various segments and partly by allowing some gases to bypass one or more segments above. A core disadvantage of this modeling approach is the assumption of chemical reaction proceeding directly towards equilibrium even if it does not reach it.

Elkem has developed two models of the Si production. The first of the two Elkem models is a Stoichiometric model, which gives a steady state approximation of the process that combines equilibrium and kinetic considerations with material and energy balances [9]. The furnace consists of two major reaction zones in which the lower one has a higher operating temperature. Furthermore, the top zone is split into two additional chambers to account for inhomogeneity in the incoming charge and the distribution of outgoing gases. Equations in the model are purely algebraic and the solution is arrived at by an iterative procedure.

The Stoichiometric model has found its application as an educational tool for process metallurgists and furnace operators. Its limitation, a steady state simulation, does not produce a clear picture of the furnace states as they evolve with time under various and continuous controlled and unforeseen disturbances.

The second Elkem model is a dynamic model based on a coupled set of nonlinear set of differential equations [8]. Formidable difficulties in numerical convergence where encountered [8]. However, further simplifications resulted in the development of the Unidimensional Dynamic Model and a solution algorithm that allowed the simulation to effectively converge to acceptable solution by the end of mid 1990's. This model has established itself as a standard for other modeling work that has followed and it has even crossed over to other process industries [15].

The Unidimensional Dynamic Model incorporates two zones in which the upper zone is vertically subdivided into a number of segments. The upper zone, referred to as the shaft, employs a relatively detailed description of the upper furnace and, hence, comprises a nonlinear set of partial differential equations. The lower zone referred to as the hearth, describes the lower furnace as a lumped system and using a non-linear set of ordinary differential equations. Both sets of differential equations are formulated based on mass and energy balances, and the shaft and hearth zones are coupled by mass and heat transfer interactions. Chemical kinetics are treated in a simple and effective manner by allowing the reaction rates to be proportional to a deviation from equilibrium and a condensed reactant concentration.

A final model known in the public domain was developed by Birger Andresen [1]. This model gives a computational fluid dynamics (CFD) description of the high temperature environment and the underlying interaction near a single submerged electrode. The model consists of partial and ordinary differential equations adapted for two-dimensional geometry. Variations in radial and vertical directions are allowed. The size of the code and the heavy computational power it requires renders this approach inefficient for any real time industrial applications at the present time.

The mathematical models described above were designed to predict and analyze the response of the furnace subject to various disturbances. Each model performs this task to some extent but none of them has been adapted to on-line process control and optimization.

The aim of the current paper is to develop a model of the Si production process, which is suitable for on-line control, data reconciliation and process optimization. We extend the model described in [8] and rewrite it so that we can use the method of lines (MOL) and finite differences to discretize the spatial derivatives of the partial differential equations. This approach yields an initial value ordinary differential equation system. Including the boundary conditions as algebraic constraints casts this former formulation into a more advantageous formulation known as the differential and algebraic equation (DAE) system. The main advantages of the DAE formulation are that it is easy to develop since the variables convey physical meaning and there is great freedom in representing the equations of the realistic process. Furthermore, the overall nonlinearity of the dynamic system may be reduced when compared with concurrent methods for nonlinear ordinary differential equations (ODEs). In a concurrent method, the algebraic constraints are eliminated [3].

The main disadvantage of the DAE approach is that specialized numerical approaches must be used. However, new solution approaches to the DAE system formulation have been developed in recent years and applied to various classes of systems. In this paper, we will show how the Si system can be adapted for the public domain DAE solver, DASPK 3.0 [4]. The solver applies a preconditioned Krylov method for solution of Backward Difference Formula (BDF) equations, which are numerical approximations to first order time derivatives.

DASPK exhibits exceptional numerical convergence and stability properties and is well suited for the solution of large-scale systems arising from method of lines discretization of partial differential equation (PDE) systems. Future work will be focused on embedding the model into an optimization package for online parameter estimation and control.

2 MODELING

2.1 The Chemical Reaction System

The overall chemical reaction that governs silicon production is:

$$SiO_2 + 2\,C \rightarrow Si + 2\,CO \qquad \text{(i)}$$

In the real system, two sets of spatially distributed reactions occur. The first set takes place in the upper cooler zone of the furnace where the rising gases react with the solid charge according to:

$$SiO + 2\,C \rightarrow SiC + CO \qquad \text{(iia)}$$

$$2a\,SiO \rightarrow a\,SiO_2 + a\,Si \qquad \text{(iib)}$$

The positive stoichiometric constant 'a' can assume an arbitrary value, for example in the vicinity of 0.08 but bounded below unity. The main reason for introducing the parameter 'a' is that the exact conversion of the species is unknown and depends on the furnace conditions. A submerged electric arc/plasma provides the heat for reaction in the lower zone:

$$\begin{aligned}(1{+}a)\,SiO_2 + SiC \rightarrow& \\ &(1{-}a)\,Si + (1{+}2a)\,SiO + CO\end{aligned} \qquad \text{(iii)}$$

The products of reaction (iii) are liquid Si metal, gaseous SiO and CO. These gases rise to the upper zone providing necessary heat and material for reactions (iia) and (iib). The configuration of the reaction scheme is cyclic in nature; each zone requires the products from the other zone in order to sustain the production of silicon metal. Unreacted SiO and CO by-product gases leave the furnace, are oxidized and CO_2 is emitted to the atmosphere while the particulate SiO_2 is captured and treated appropriately. The silicon metal is recovered or *tapped* from the furnace during timed intervals, giving the process a semi-batch nature. To maintain an even distribution of reactants and eliminate any undesired gas cavities, the furnace is stoked. Stoking is imposed on the system by intermittently mechanically pressing and breaking the partially reacted crusted charge downward using a paddle type device.

We propose to model the present chemical system utilizing three independent reactions:

$$SiO_{(g)} + 2\,C \leftrightarrow SiC + CO_{(g)} \qquad \text{(iv)}$$

$$2\,SiO_{(g)} \leftrightarrow Si + SiO_2 \qquad \text{(v)}$$

$$SiO_{(g)} + SiC \leftrightarrow 2\,Si + CO_{(g)} \qquad \text{(vi)}$$

Temperatures in upper furnace decrease with decreasing furnace depth. The relatively less endothermic reaction (iv) will predominate there, converting feed C to SiC, which is gradually transported downward to the lower furnace. A few SiO condensation reactions are believed to take place in the furnace but due to their comparatively slow rates, only reaction (v) is considered in the furnace. In the vicinity of the lower furnace, reaction (vi) begins to compete due to the protruding thermal effect of the electrodes.

In the lower furnace, the temperatures are much higher and a vertical thermal gradient is considered to be minimal. The highly endothermic reaction (vi) will predominate. Due to the elevated thermal environment, reaction (v) will now reverse, reacting SiO_2 and Si to produce SiO gas. Based on the current state of the furnace, equilibrium constraints determine the extent of each reaction.

The reaction system presented is currently a sufficient description if a goal of adequacy and simplicity is to be achieved in modeling the substantially complex silicon process. Properly formulated kinetic models based on some given deviation from equilibrium provide a satisfactory number of degrees of freedom for fitting the parameters in order to adapt the model to different furnace configurations scattered throughout industrial plants.

2.2 Chemical Kinetics

The rate of each reaction is defined as being proportional to the concentration of the condensed reactant involved and an Arrhenius temperature-depended factor. An additional driving force is expressed as a deviation from the equilibrium pressure, which is assumed to be a ubiquitous 1 atm. As an example, a complete rate expression for reaction (iv) has the form:

$$R_{iv} = a_{iv}\, c_C\, \Delta p_{iv}\, e^{(-E_{iv}/RT)} \qquad (2.2.1)$$

where R_{iv} is the reaction rate, a_{iv} is the proportionality factor, c_C is the local concentration of carbon, Δp_6 is the deviation from equilibrium pressure, E_{iv} is the Arrhenius factor activation energy, R is the molar gas constant and T is the local temperature.

Ideally, all kinetic parameters such as activation energies (E) and proportionality/rate constants (a_{iv}) should be experimentally determined for each reaction rate expression. Unfortunately, such data are limited and/or unavailable. To compensate, parameters are adjusted until the model satisfactorily matches a given furnace behavior. To diminish ambiguity and complexity in parameter fitting, the Arrhenius term is neglected by setting the activation energies (E) to zero for all rate expressions in our preliminary studies.

2.3 Model Equations

The furnace is modeled with a coupled set of nonlinear partial differential equations. The solid components include SiO_2 (feed quartz), C, SiC and condensed Si and SiO_2. Liquids include Si and SiO_2, and the gases are CO and SiO. Halvorsen [8] proposed to define an extended species as "SiO", a condensate mixture of SiO_2 and Si, for the solid and liquid phases generated by forward reaction (v). The present work departs from this formulation and explicitly treats the condensate species for solid and liquid phases as part of the overall species in respective phases, SiO_2 and Si. Flux velocities are defined as positive for gases and negative for solids and liquids. Assuming that the species of each aggregate state move at a common speed, the mass balances are:

$$\frac{\partial c_i}{\partial t} + \frac{\partial (v_S c_i)}{\partial z} = \sum_j r_{ij} R_j - S_i \qquad \mathrm{i = C, Si, SiC, SiO_2} \qquad (2.3.1)$$

$$\frac{\partial l_i}{\partial t} + \frac{\partial (v_l l_i)}{\partial z} = \sum_j s_{ij} R_j + S_i \qquad \mathrm{i = Si, SiO_2} \qquad (2.3.2)$$

$$\frac{\partial g_i}{\partial t} + \frac{\partial (v_g g_i)}{\partial z} = \sum_j q_{ij} R_j \qquad \mathrm{i = CO, SiO} \qquad (2.3.3)$$

where t is time, z is the vertical shaft location, c_i, g_i, l_i are the local concentrations of solids, liquids and gases respectively, v_S, v_l, v_g are the velocities for solids, liquids and gases respectively, R_j is the reaction rate for reaction j, S_i is the melting rate for component i and r_{ij}, s_{ij}, q_{ij} are stoichiometric coefficients in reaction j for component i.

Heats of reaction are not explicitly included in the energy balance equation since enthalpies with elemental bases are chosen. If temperature T were to be chosen then the reaction enthalpies would appear explicitly. The energy balance equation is:

$$\frac{\partial H}{\partial t} + \frac{\partial (v_s H_s)}{\partial z} + \frac{\partial (v_l H_l)}{\partial z} + \frac{\partial (v_g H_g)}{\partial z} = \frac{\partial}{\partial z}\left(\lambda \frac{\partial T}{\partial z}\right) \qquad (2.3.4)$$

where H is the total enthalpy, H_S, H_l, H_g are the sums of enthalpies for solids, liquids and gases respectively, and λ is the heat conductivity.

Complementary equations expressing reaction rates for reaction j and involving a condensed reactant i based on deviation from equilibrium and defined positive for the forward direction are:

$$R_j = a_j\, c_i\, \Delta p_j\, e^{(-E_j/RT)} \qquad (2.2.1)$$

The phase transition from solid to liquid for SiO_2 is not sharply defined and a melting rate equation for this specie is introduced, retaining only one liquid velocity in the PDE:

$$S_i = \begin{cases} 0 & T < T_{mi} \\ a_i (T - T_{mi}) & T \ge T_{mi} \end{cases} \qquad \mathrm{i = SiO_2} \qquad (2.3.5)$$

where a_i is the melting coefficient for the specie, T is the local temperature, T_{mi} is the melting point temperature for the specie.

In this study, we are only concerned with modeling the furnace in a continuous regime; hence, stoking cycles are ignored. Continuous operation assumes that the voids created by reactions; melting and transport of material to the hearth are immediately filled by adding cold charge at the top of the furnace. For this purpose, additional equations for computation of the phase velocities are required and are supplemented by deriving a transport equation for the volume fraction occupied by solid C, Si, SiC and SiO_2, treating the remaining species as interstitials. The solid velocity equation is:

$$\frac{\partial v_s}{\partial z} = -\frac{V_{SiC}R_{vi} - (V_{Si} + V_{SiO_2})R_v + V_{Si}S_{Si} + V_{SiO_2}S_{SiO_2}}{f} \quad \mathrm{i = C,\, Si,\, SiC,\, SiO_2} \qquad (2.3.6)$$

$$f = \sum_i f_i = \sum_i V_i c_i$$

V_i are the molar volumes for C, Si, SiC, SiO_2, f_i is the volume fraction occupied by C, Si, SiC, SiO_2

Considering only solid phase dynamics, liquid and gas concentration time derivatives are ignored and the equation for these reduce to zero:

$$\frac{\partial l_i}{\partial t} = 0 \quad \mathrm{i = Si,\, SiO_2} \qquad (2.3.7)$$

$$\frac{\partial g_i}{\partial t} = 0 \quad \mathrm{i = CO,\, SiO} \qquad (2.3.8)$$

The nonlinear terms in equations 2.3.2 and 2.3.3, namely $v_l l_i$ and $v_g g_i$, become numerically coupled and are formally referred to as Q_i and G_i, respectively. In light of these modifications, additional equations are now required since independent values of v_l and v_g are required in (2.3.4).

An alternative equation is obtained by rewriting the liquid enthalpy term based on a specific enthalpy for each component. For the flux gases, an assumed ratio between them is utilized to calculate the coupled terms. These are reflected in the equations below:

$$v_l H_l = v_l \sum_i l_i h_i(T) = \sum_i (v_i l_i) h_i(T) \quad \mathrm{i = Si,\, SiO_2} \qquad (2.3.9)$$

$$\frac{p_{SiO}}{p_{CO}} = \frac{v_g g_{SiO}}{v_g g_{CO}} \qquad (2.3.10)$$

The heat transfer between particles in a hot packed bed is effectively transferred via radiation. Appropriate expressions for this phenomenon have been reported by Schotte [16] and are employed in the model:

$$Q_r = \left(\frac{1-\delta}{\frac{1}{\lambda} + \frac{1}{k_r{}^\circ}} + \delta k_r{}^\circ \right) \frac{\pi}{4} D_p^2 \frac{1}{1-\delta} \frac{dT}{dz} \qquad (2.3.11)$$

$$k_r{}^\circ = 0.692 \varepsilon D_p \frac{T^3}{10^8}$$

where δ is the particle void fraction, D_p is the particle diameter, ε is the emissivity, λ is the thermal conductivity and $k_r{}^\circ$ is the radiation contribution to the thermal conductivity of the bed.

The initial and boundary conditions essential to fully specifying the model are derived from the inlet and outlet furnace operating conditions. Solid components, C and SiO_2 comprise the inlet materials, and gases CO and SiO and liquid Si metal comprise the outlet materials. Solid concentrations, C and SiO_2, are specified at the top. Zero concentrations of solid Si and SiC are specified at the furnace top (L). Production rates for liquid fluxes of Si (and slag SiO_2) are specified to have zero value at the top of the furnace. CO gas flux of zero is specified at the furnace bottom, while SiO gas flux is specified at the initial condition value. A solid velocity of zero is specified at the furnace bottom.

$$\begin{array}{lll} c_C(t, L) = IC\ value & c_{SiO2}(t, L) = IC\ value & (v_g g_{CO})(t, 0) = 0 \\ c_{Si}(t, L) = 0 & (v_l l_{Si})(t, L) = 0 & (v_g g_{SiO})(t, 0) = IC\ value \\ c_{SiC}(t, L) = 0 & (v_l l_{SiO2})(t,L) = 0 & v_s(t, 0) = 0 \end{array} \tag{2.3.12}$$

Our current model development efforts allowed us to successfully build a material balance-based process model. All energy and thermal effects have been kept to the minimum and postponed until this approach could be verified with appropriate testing. A fixed temperature profile believed to reflect the thermal conditions of the furnace has been constructed and used in lieu of the energy balance. Any further reference to the energy balance is omitted.

3 SOLUTION STRATEGY

3.1 PDE Solution Strategy Utilizing Method of Lines Approach

Various numerical techniques exist for obtaining solutions to various classes of partial differential equations (PDEs). Finite difference, finite volume and finite element methods are among the most popular [14], and we focus our intentions on utilizing a finite difference scheme, specifically the method of lines (MOL).

A general concept of a finite difference scheme is to algebraically approximate the spatial and temporal derivatives with algebraic differences in the dependent variable's discrete value. A grid is constructed by discretizing the spatial domain of the problem at hand into finite but small sections. A line slope of neighboring grid nodes constructs a local approximation for the spatial derivative. Time derivative are treated similarly. Difference formulas are employed to construct an approximation relative to a current node. Enumerating this procedure for all grid nodes fabricates a set of algebraic equations, which can be solved with any standard linear solver [14].

Method of lines approach is a modification of the finite difference scheme by means of only requiring a spatial discretization of the PDE system. The time derivatives are retained and a set of ordinary differential equations (ODEs) emerges, which can be solved with a standard ODE solver.

Furthermore, any additional algebraic equation and boundary condition formulated as an algebraic equation explicitly not substituted into the ODE set, constructs a *differential and algebraic equation* (DAE) system [3]. Specifically, the method of lines approach applied to the solution of PDEs generates a *semi-explicit DAE* system [3]. In the next section, we present some underlying concepts concerning general DAE systems.

Several important numerical issues arise when considering finite differences applied to first order hyperbolic PDEs. These equations are particularly difficult to solve as they transmit discontinuities without dispersion or dissipation [6]. Moreover, the finite difference approach introduces dispersion and numerical oscillation producing potentially detrimental effect in the context of solution accuracy and stability. One remedy to overcome this, at least with minimal effects, is to employ directional or upwind/downwind finite differences over central finite differences. Furthermore, including more terms in the computation of the derivatives at a node increases the order (accuracy) and reduces the stiffness of the resulting ODE set [10].

3.2 Differential and Algebraic Equations

Only recently has there been an increased interest in directly working with differential and algebraic equations (DAE) as opposed to ordinary differential equations (ODEs). Generally, an ODE system can be written as an implicit first order system:

$$F(t, y(t), y'(t)) = 0 \tag{3.2.1}$$

where F and y are vector valued. A typical reformulation of this into an explicit or normal form

$$y'(t) = f(t, y(t)) \tag{3.2.2}$$

is the basis for most classical numerical integration schemes. The procedure of converting (3.2.1) to (3.2.2) involves substituting the algebraic constraints into the differential equation. This is not always possible to do since the explicit expression may not be available in analytical form. The equations to integrate may also become numerically complex.

A shift away from dealing with (3.2.2) and working directly with (3.2.1) has been widely adapted due to numerous advantages that are inherent to the original formulation. Numerical techniques have been developed by Gear [7] and Petzold [3].

Many times equation (3.2.1) may not even have an explicit reformulation. The constraints may appear explicitly as:

$$F(x', x, y, t) = 0 \tag{3.2.3a}$$

$$G(x, y, t) = 0 \tag{3.2.3b}$$

where the Jacobian of F with respect to x' (denoted $F_{x'}$) is nonsingular or $F_{y'}$ in (3.2.1) is singular. Numerical methods to solve (3.2.3) have not been available until recently.

There are a few major advantages of working directly with (3.2.1), rather than trying to rewrite it as an explicit ODE. First, when the physical problems are simulated, the model often takes the form of a DAE depicting a collection of physical relationships between algebraic variables and some of their derivatives. In case of nonlinear models, it maybe time consuming or even impossible to reformulate the problem into an explicit ODE (3.2.2), and if it is accomplished, the variables may lose their physical meaning. Secondly, changing parameter values can alter the relationship between variables and require different explicit models accompanied by a different solution. Given that the original DAE can be solved directly, the analysis of modeling and parameter modifications effects is greatly facilitated. From a numerical perspective, reformulation, if possible, can lead to a destruction of sparsity and prevent exploitation of the system structure.

Differential algebraic equations arise for various classes of problems, and most of the time, these categories overlap to some extent. The type of problem is based on the method the equations are derived by, rather than on the type of resulting equations. In the framework of this study, we are concerned with DAE systems that arise from a method of lines discretization of partial differential equations (PDEs), which yield a semi-explicit DAE system.

The advantages of using a method of lines approach are twofold. First, it is computationally efficient; the DAE software, which is developed to be robust and computationally efficient, assumes the burden of time discretization and of appropriately choosing the time steps. Hence, the stability and accuracy of an evolving solution is maintained. Second, the analyst must only be concerned with discretizing the spatial derivatives and not be hampered with writing dedicated computer code to solve the PDE system. In light of the present discussion along with the previous modeling work, it is a natural choice to proceed with formulating the nonlinear dynamic model based on the method of lines/differential and algebraic equation approach.

3.3 Solution Method

The basic idea for solving a DAE system originated from Gear's procedure for replacing the time derivative in *(3.2.1)* with a backward difference formula (BDF):

$$F(t_n, y_n, \rho y_n/h_n) = 0 \tag{3.3.1}$$

where subscript n represent a time step, $h_n = t_n - t_{n-i}$, and $\rho y_n = \Sigma \alpha_i y_{n-i}$, α_i's are the BDF coefficients for i = 0, 1, …k. The equations that arise with this transformation are nonlinear and a modified Newton method is utilized for the solution at iteration *m*:

$$y^{m+1} = y^m - c(\alpha F_{y'} + F_y)^{-1} F(t, y^m, \alpha y^m + \beta) \tag{3.3.2}$$

where α and β are constants that are evaluated at t_n.

At each Newton iteration m, a linear system of the form $Ax = b$ arises, where

$$A = \alpha F_{y'} + F_y \tag{3.3.3a}$$

$$x = y^{m+1} - y^m \tag{3.3.3b}$$

$$b = -cF(t, y^m, \alpha y^m + \beta) \tag{3.3.3c}$$

Specialized theory, numerical techniques and software for the solution of a DAE system have been developed and pioneered by numerous researchers, i.e. Petzold and co-workers. The first generation of the software package called *DASSL* [3], developed in 1982, is intended to solve DAE systems of index zero and one only. Higher index DAEs present problems to the code by jeopardizing stability, time stepping procedure and convergence tests. In most cases, including this one, problems originating from method of lines are index one.

The second generation of DAE solvers is the DASPK, recently developed by Petzold *et al.* [4]. This software package is specifically designed to handle large-scale systems such as DAE systems arising from the method of lines discretization of PDEs. In addition to performing identically to DASSL, it includes an *iterative option* that, if applied to highly non-linear DAE systems, outperforms the DASSL's *direct method.*

The direct method approach can be handled by DASSL and DASPK, and the linear system (3.3.3) is solved by dense direct or banded direct Gaussian elimination with partial pivoting via LINPACK. The iterative method solves the linear system using a *preconditioned Generalized Minimal Residual (GMRES) iterative method*, a Krylov subspace projection method that solves the system (3.3.3) inexactly.

One requirement of the iterative method is provision of a preconditioning matrix P. This transforms the original system into one that can be easier to solve

$$(P^{-1}A)x = P^{-1}b \Rightarrow A^{*}x = b^{*} \tag{3.3.4}$$

providing that *(i)* linear system $Px = c$ can be solved economically, and *(ii)* P is close to A. Condition *(i)* is essential since performing this on $A^{*}x = b^{*}$ requires evaluating vectors of the form $P^{-1}c$. Condition *(ii)* is less well defined, but convergence for a transformed system is better since it is closer to the identity matrix for which convergence is immediate.

In developing the preliminary material balance-based model, we utilized the direct method of DASPK for the solution of the governing equations. The results are satisfactory and will be presented next.

4 PRELIMINARY MODEL RESULTS

At the present stage, we have successfully developed a material balance-based process model and incorporated an estimated temperature profile in lieu of the energy balance. DAPSK solver is coded in Fortran programming language and consequently our model is developed in Fortran and in accord with the DASPK documentation.

A set of consistent initial and boundary conditions (ICs) is required by the solver in order to successfully integrate the system. ICs for the dynamic variables were inferred from the Unidimensional Dynamic Model simulation results and implemented in the model. DASPK includes an option that calculates the remaining ICs for the algebraic variables based on the ICs of the dynamic variables, *i.e.,* $v_l l_{Si}$ formally referred to as Q_{Si} versus c_C. We have utilized this option and produced a full set of ICs for the problem. The boundary conditions were then assigned by fixing the initial value of each variable at a proper furnace location (top or bottom) ensuring consistency.

Originally, we have discretized the first order spatial derivatives employing second order central differences (4.1) [2]

$$\left.\frac{\partial u}{\partial x}\right|_i = \frac{u_{i+1} - u_{i-1}}{2\Delta x} + O(\Delta x^2) \tag{4.1}$$

It was realized that this approach produced intensifying oscillation and instabilities in the state profiles as system was integrated for longer times. As a result, a combination of directional (upwind/downwind) differences was incorporated for discretizing the first order spatial derivatives. Namely, a forth order unbiased upwind difference (4.2) and a forth order biased upwind difference

(4.3) [2], biased meaning upwind with a central-like

$$\left.\frac{\partial u}{\partial x}\right|_i = \frac{11u_i - 18u_{i-1} + 9u_{i-2} - 2u_{i-3}}{6\Delta x} + O(\Delta x^4) \tag{4.2}$$

$$\left.\frac{\partial u}{\partial x}\right|_i = \frac{3u_{i+1} + 10u_i - 18u_{i-1} + 6u_{i-2} - u_{i-3}}{12\Delta x} + O(\Delta x^4) \tag{4.3}$$

influence, were combined to integrate the system with minimal oscillations and dispersions.

Two sets of simulation case studies were prepared utilizing a spatial discretization level of 80 nodes. In simulation 1, the system was subsequently integrated over 5000 seconds at the resolution of fifty (50) 100 second time horizons (75 CPU seconds on a 933MHz Intel Pentium III). In simulation 2, the system was subsequently integrated over 3000 seconds at the resolution of thirty (30) 100 second time horizons (78 CPU seconds on a 933MHz Intel Pentium III). In simulation 1, the first order derivatives were approximated with second order central finite differences while simulation 2 utilized a combination of forth order biased and unbiased upwind differences. Simulation 1 (Figure 1) ~~profiles exhibit strong oscillation; specifically the~~ QSi profile intensifies in these oscillations nearly over the entire furnace length. On the other hand, simulation 2 (Figure 2) profiles exhibit impressive smoothness.

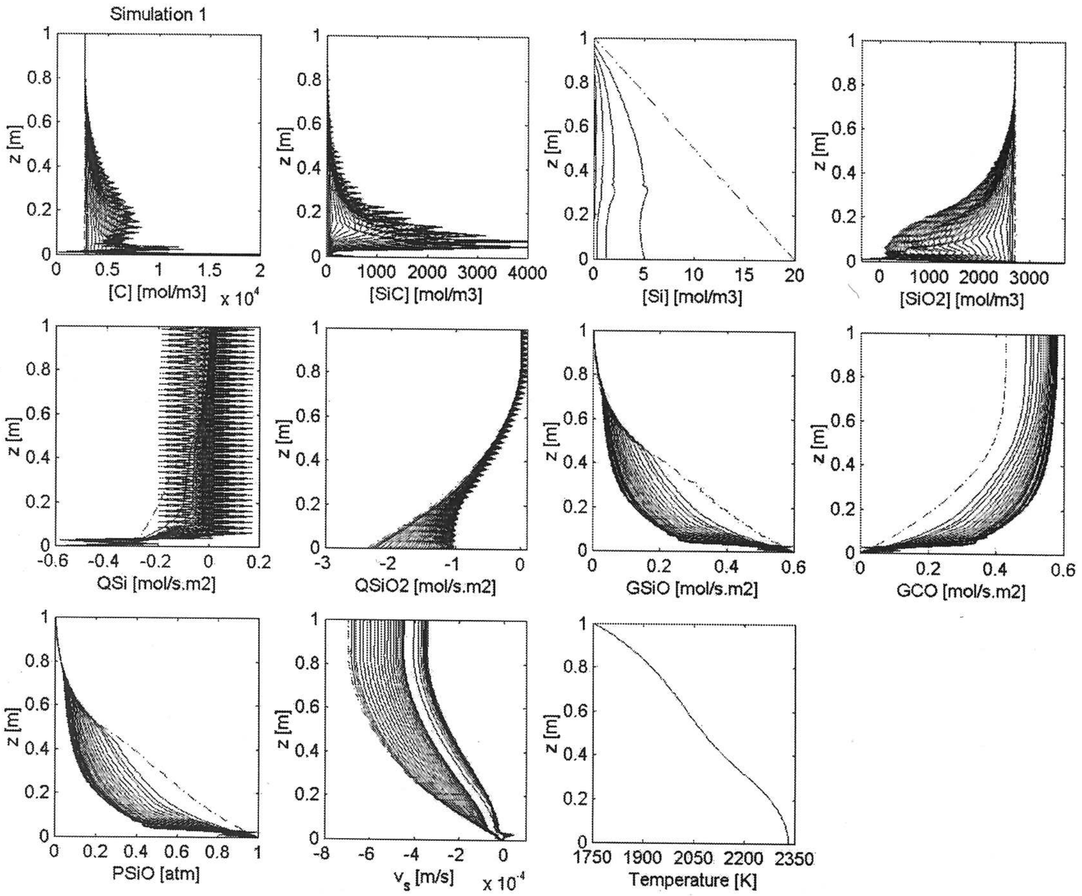

Figure 1. Simulation 1 state profiles. Dashed line represent IC profile and subsequent profiles represent dynamic evolution. Second order central finite difference approximations were utilized to discretize the spatial derivatives.

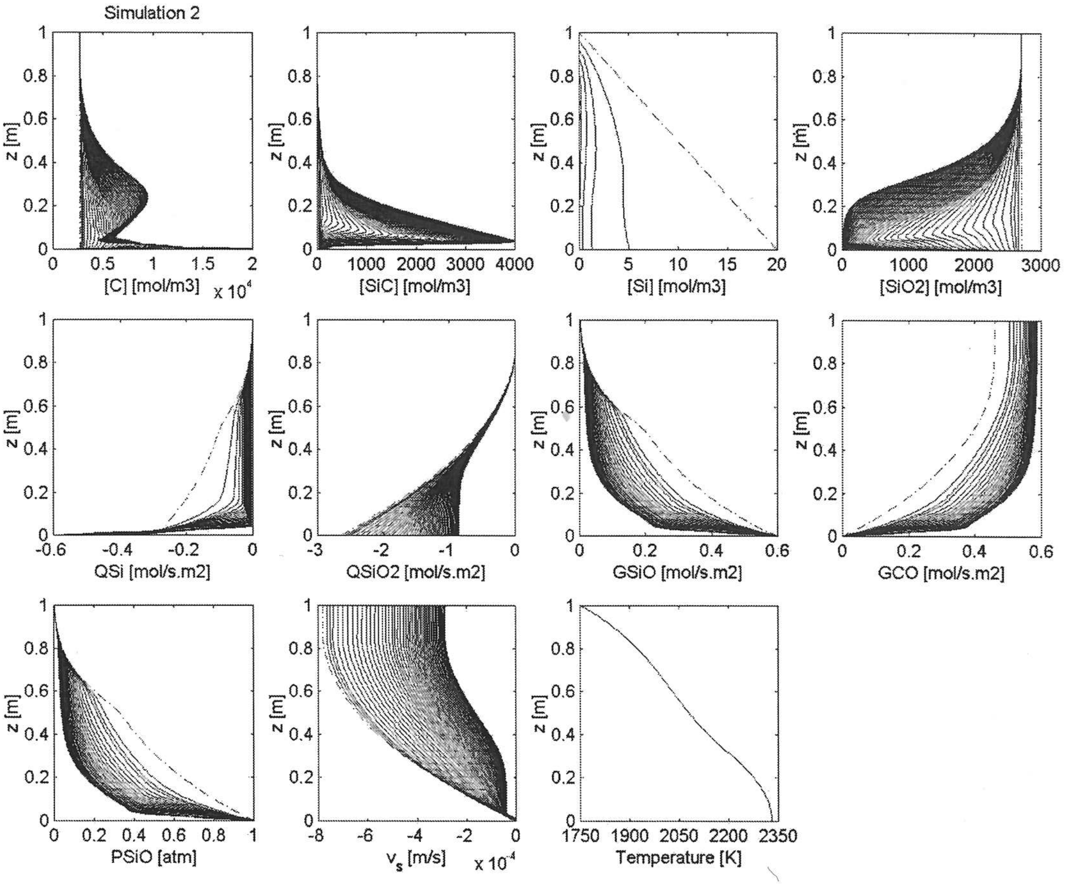

Figure 2. Simulation 2 state profiles. Dashed line represent IC profile and subsequent profiles represent dynamic evolution. Forth order biased and unbiased upwind finite difference approximations were utilized to discretize the spatial derivatives.

5 CONCLUSIONS AND FUTURE WORK

The preliminary material balance based process model was developed to assess the suitability of the solution strategy, the DAE formulation approach and performance of the DASPK solver. Case studies reflect that the assumptions made are correct and further development is currently ongoing. Integration of the energy balance will be considered next and achieved by methods outlined. The methods for incorporating discrete time events such as charging, stoking and tapping represent the next challenge and hence a final step of the development stage. The initial and boundary conditions representing realistic furnace behavior will be generated from plant data supplied by Elkem. With a properly functioning process model, additional elements required by the DASPK solver utilizing the iterative method will be constructed, namely an approximation of the Jacobian and a preconditioner. Completing these requirements will allow the model performance to be assessed.

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