Model-Based Optimization of Industrial Gas-Solid Reactors†

Venkataramana Runkana
1 Tata Research Development and Design Centre, Tata Consultancy Services Limited, India

Abstract

Heterogeneous non-catalytic gas-solid reactors are commonly used in the production of chemicals, metals and metal oxides, for example, rotary drum reactor for producing quick lime; blast furnace, a moving bed reactor for producing iron, etc. Industrial processes involve multicomponent mixtures of solids as most of the naturally occurring materials such as minerals are multicomponent in nature. Generally the raw materials are heterogeneous and their physical and chemical characteristics vary from one source to another. The solid particles are also usually moist, the moisture content varying from season to season. The physico-chemical phenomena that take place in these reactors include flow of gases through porous media, heat transfer between the gases, solids, equipment and the environment, evaporation and condensation of moisture, reactions between gases and solids and within a single phase, melting and solidification depending on the temperatures inside the reactor, etc. The flow of particles is also of great importance especially in rotary drum and moving bed reactors. General features of the mathematical models for non-catalytic reactors are described along with models for the phenomena mentioned above. Case studies from the iron and steel industry on model-based optimization of production of direct reduced iron in a rotary kiln and induration of wet iron ore pellets on a moving packed bed reactor are discussed here.

Keywords: multiphase reactors, modeling, optimization, soft-sensors, iron and steel, pelletization

1. Introduction

Production of many commodity chemicals, metals, ceramics and polymers involve reactions between gases and solid particles, which may act as catalysts in some cases and in other cases, participate in the reaction with the gases directly. The reactors commonly used in the industry include stationary and moving packed bed reactors, rotary drum reactors or rotary kilns, vertical shaft kilns or moving bed reactors, fluidized bed reactors, rotary hearth furnaces, etc. For example, vertical shaft or rotary kilns are used for the production of quick or burnt lime in the soda ash, pulp and paper (Fernandes and Castro, 2000), and iron and steel industries (Watkinson and Brimacombe, 1982; Davis and Englund, 2003; D’Abreu et al., 2008). Rotary kilns are also used for producing sponge iron or direct reduced iron (Feinman and Mac Rae, 1999), cement (Peray, 1984; Mastorakos et al., 1999; Majumdar et al., 2007), for waste incineration (Marias, 2003; Yang et al., 2003), and for combustion or pyrolysis of coal (Patisson et al., 2000a, 2000b; Li et al., 2003), maize (Klose and Weist, 1999), etc. Moving packed bed reactors are employed for sintering of iron or zinc ores (Chatterjee et al., 1993; Nakano et al., 2010; Ahn et al., 2013), for induration of iron ore pellets on a moving or traveling grate or strand (Thurlby et al., 1979; Barati, 2008; Sadrnezaad et al., 2008), and for gasification of coal (Yoon et al., 1979; Hobbs et al., 1992). The blast furnace, a moving bed reactor, is utilized for making hot metal or liquid iron in an integrated steel plant (Omori, 1987; Ueda et al., 2010a). Fluidized bed reactors are employed for roasting of zinc and sulfide ores (Nyberg, 2004; Shu et al., 1999), for the production of polycrystalline silicon (Ranjan et al., 2011) and polyethylene (Kiashemshaki et al., 2006), etc. Rotary hearth furnaces are employed for reduction of iron ores by volatiles (Sohn and Freuhan, 2005, 2006a, 2006b). In several cases, besides gases and solids, the reactor too is non-stationary. For example, the rotary kiln revolves around its axis whereas sintering (see Fig. 1) or induration reactors move continuously on a traveling grate. The industrial scale reactors are also generally large in size because of the large tonnage production of materials. For example, the reactors used for sintering of wet iron or zinc ore granules and for induration of wet iron ore pellets are rectangular in shape and have volumes in excess of 500 m³ whereas the blast furnace is irregular in shape and has volumes in excess of 5000 m³ (Ueda et al., 2010a). The focus of this paper is on the non-catalytic gas-solid

† Received 7 October 2014; Accepted 14 November 2014
J-STAGE online 28 February 2015
1 54-B, Hadapsar Industrial Estate, Pune, India, 411013
E-mail: venkat.runkana@tcs.com
TEL: +91-20-66086268 FAX: +91-20-66086399
The process engineers face several challenges in process monitoring, optimization and control. Firstly, as the earth’s natural resources are getting depleted, availability of good quality raw materials from a single source has become relatively rare. Hence, the plant operators have to blend raw materials from different sources which invariably leads to inhomogeneity and variability in quality. Secondly, the production as well as product quality demands keep changing depending on the market demands. Thirdly, as the effect of industrial operations on climate change is well established, there is a need to optimize the operations to ensure environmental compliance.

It is evident that one needs to undertake multi-objective optimization in order to achieve these goals. However, because of difficulties or uncertainties with numerical convergence and because it is computationally intensive, single objective optimization is commonly carried out to achieve the highest priority goal while ensuring that the variables representing the other objectives are within the desired limits, as stipulated by either upstream or downstream process requirements.

In the case of industrial process optimization, there are stringent requirements for the mathematical models. The key requirements of these models and related software tools include validity over diverse operating conditions, stringent prediction accuracy, ability to handle abnormal or missing values of variables, interactions between different input variables, uncertainty in input data, error and uncertainty of model predictions within and beyond the operating regimes for which the model was tuned or customized, etc. This necessitates development of reasonably comprehensive and accurate models, such as those mentioned above, and testing their validity and accuracy with current and past data. This, in turn, requires extensive application of data mining and analysis techniques, identification of key process variables from a large set (consisting of several hundreds of variables in some cases), and development of relationships between variables. One of the important steps here is the prediction of variables that cannot be measured online, using fundamental models and establishing relationships between measured and unmeasured variables using data collected over several years. Hence, there is a need for high fidelity lumped or distributed parameter models which could be deployed as soft-sensors for on-line optimization as well as for advanced process control. Moreover, these models could be incorporated into general purpose simulation software tools such as SolidSim for processing of particulate solids (Hartge et al., 2006) for designing processes.

### 1.2 Need for soft-sensors and data analytics

It is difficult and in some cases impossible to measure many key intrinsic process variables either because of lack of suitable sensors or instruments or because the particles and/or the equipment may be in motion which makes it difficult to make measurements. The sensing element may also get damaged due to the impact of solids. Moreover, quality parameters of products that are in particulate form, for example, strength, reactivity or chemical analysis, have to be determined through off-line laboratory analyses as it is not possible to determine them in-line in most cases. Hence, there is a necessity to develop and install soft-sensors which could be employed for on-line optimization and control.

As is the case with most of the manufacturing processes which involve complex interplay of several physico-chemical phenomena, it is always not feasible to represent all the phenomena using first principles and relate them to properties of the end-products. For example, the strength of iron ore sinter and pellets depend strongly on the mineral phases formed and reactions that occur at high temperatures but a representative model for these phenomena is still elusive. In such cases, the usual practice is to resort to data-based modeling techniques (Ge et al., 2014; Kaneko and Funatsu, 2014) for soft-sensor development. However, it would be more appropriate to address this problem using a hybrid phenomenological-statistical modeling approach, employing the mathematical model of the process to predict internal condition of the reactors, for example, temperature, chemical composition, etc., and then use the...
Gas-solid reactors are very common in several industries and fundamentals of reaction kinetics and reactor design have been described in detail elsewhere (Szekely et al., 1976; Smith, 1981; Levenspiel, 1988). Modeling and optimization of gas-solid reactors has been studied extensively (Schaefer et al., 1974; Cumming and Thurby, 1990; Rovaglio et al., 1994; Cross and Blot, 1999; Sadri et al., 2007) and it is difficult to review the entire subject in detail here. The author has been involved in the development of mathematical models for industrial scale gas-solid reactors, related software tools (Virtual Sinter™, Virtual Indurator™ and DRIKS™) and their application for various industrial problems, for example, sintering of iron ore fines (Venkataramana et al., 1997, 1998, 1999, 2002; Gupta and Runkana, 2000; Kapur and Runkana, 2003), induration of iron ore pellets on a traveling grate (Majumder et al., 2009, 2012; Mitra et al., 2009; Cavalcante et al., 2012; Runkana and Majumder, 2013), direct reduction of iron ore by coal in a rotary kiln (Runkana et al., 2007; 2010a, 2010b, 2010c), and calcination of limestone in a vertical shaft kiln, etc. The objective here is to share this knowledge of model development incorporating important physico-chemical phenomena, simulation and optimization techniques employed and details of their demonstration for a couple of industrial cases. However, appropriate references are provided so that the interested reader can explore the subject in greater detail.

This paper is organized as follows: Mathematical modeling of gas-solid reactors is described in detail in Section 2. The simulation and optimization techniques applied are discussed in Section 3. Industrial case studies on direct reduction of iron ore by coal in a rotary kiln and induration of iron ore pellets on a traveling grate are presented in Sections 4 and 5, respectively. Finally, concluding remarks and suggestions for future research are provided in Section 6.

2. Mathematical modeling of gas-solid reactors

The industrial reactors could be co-current, cross-current or counter-current in nature with respect to the flow of gases and solids. The process is heterogeneous in nature since particulate solids are involved and it is appropriate to represent gas-solid reactors using distributed parameter models. However lumped or distributed parameter models are formulated, depending on the intended application of the model. Mass and energy balance models are formulated as nonlinear algebraic equations, especially in the preliminary stages of development, to determine unmeasured process variables and for data reconciliation. The one-dimensional models are usually formulated as a set of ordinary differential equations whereas two- and three-dimensional models are formulated as partial differential equations employing the Cartesian or cylindrical coordinates, depending on the geometry of the reactor.

The basic physico-chemical phenomena that take place in gas-solid reactors include: charging of particles and formation of particulate bed, flow of particles and gases and their mixing, heat transfer between gases, solids, equipment and the environment, mass transfer between gases and solids, drying or evaporation of moisture from wet particles or agglomerates, reactions within or between gas and solid phases, partial melting at high temperatures and subsequent solidification as temperature decreases, condensation of moisture from the gas phase, etc. While the individual phenomena occur based on the thermodynamics or kinetics that govern them, there will be interactions amongst the phenomena that make the process complex to understand. Because of the interactions amongst the underlying phenomena, it is important to incorporate all the representative phenomena, to the extent possible, and solve the equations in a coupled manner.

From the process systems engineering perspective, the gas-solid reactors are represented in terms of process variables (flow rate, chemical composition, temperature, pressure, particle size distribution, etc.), raw material characteristics (chemical composition, particle size distribution, surface roughness, reactivity, strength, etc.), equipment or reactor design parameters (length, diameter, height, type of refractory, refractory wall thickness, speed of revolution, traveling grate speed, material used for shell and shell thickness, etc.), environmental conditions (pressure, temperature, humidity, chemical composition), product quality indices (strength, reactivity, porosity), physical or thermodynamic properties of the materials involved as functions of temperature and pressure (density, viscosity, specific heat, thermal conductivity, etc.), and model parameters (heat and mass transfer coefficients, reaction rate constants, etc. as functions of temperature and/or pressure).

Porosity of individual particles or agglomerates and the particulate bed voidage play an important role in the heat and mass transfer between the gases and the solids. The bed voidage especially is a crucial parameter and depends on the size and shape distribution of particles, deformation and consolidation behavior in the case of agglomerates, reactor geometry, and on the mechanism of charging of the particles into the reactor.

2.1 Charging and flow of solids

Permeability of the particulate bed has a strong influence on the key performance indicators of the reactor, namely, productivity, product quality, energy or fuel effi-
The flow of particles inside the reactor depends on the reactor geometry, size and shape distributions of the particles and the fluid flow, especially in the case of fine particles and fluidized bed reactors. Kunii and Levenspiel (1997) described a number of gas-solid contacting patterns depending on the gas velocity and bed structure in a fluidized bed. The flow of heterogeneous particulate solids in rotary drums was analyzed in detail by Abouzeid and Fuerstenau (2010) along with experimental evidence of the effect of particle properties on residence time distribution inside the drum.

Since the computational time required for simulation of individual particles is enormous, the continuum approach was followed earlier to simulate flow of particles. Saeman (1951), and Kramers and Croockewit (1952) first developed a steady state model for transport of solids. Saeman (1951) used experimental observations of Sullivan et al. (1927) to develop a mathematical expression for solids residence time and volumetric transport rate in the kiln as a function of kiln rotation speed and its angle of inclination. Kramers and Croockewit (1952) developed an expression for bed height variation along the kiln axis as a function of volumetric flow rate of solids, kiln inclination, angle of repose of solids, kiln internal radius and speed of rotation. Henein et al. (1983) developed a diagram to represent several modes of operation of a rotary kiln such as slipping, slumping, rolling, cascading, cataracting and centrifuging. Boateng and Barr (1996) developed an analytical model to predict particle segregation within a rotary kiln as a function of diameter and speed of rotation of kiln and bed depth. Khakhar et al. (2001) developed a continuum model to predict the extent of mixing and size segregation for binary mixtures of different sized particles but of same density. Liu et al. (2005) developed an analytical expression to calculate maximum active layer thickness and mass flow rate in the layer as function of kiln geometry and operating parameters.

With the advent of computationally efficient processors and high performance computing systems, the DEM is also employed nowadays for particle flow simulations. Moysey and Baird (2009) studied segregation of Nickel pellets in the surface flow of a packed bed whereas Liu et al. (2013) applied DEM for studying the transverse mixing of wet particles in a rotary drum. Mio and coworkers (2008, 2009) studied the effect of chute angle on charging behavior of sintered iron ore in bell-less type charging system of a blast furnace and validated simulations of particle segregation on the chute with laboratory experimental data. Ueda et al. (2010b) simulated descent of particles and its effect on the stress distribution within the packed bed of a blast furnace. Discrete element simulations for particulate systems and their industrial applications were reviewed in detail elsewhere (Zhu et al., 2008).

The structure of the particulate bed that forms inside the reactor depends quite a lot on the mechanism of charging, the device used, and segregation behavior and physical properties of the particles. For example, chutes or inclined plates are used for charging of particles from silos or bins onto the moving grate in the case of sintering or inclined plates are used for charging of particles from vertical shaft kilns and blast furnaces (Omori, 1987; Mio et al., 2008, 2009). Since particles are heterogeneous in nature, segregation is a common phenomenon. It could happen due to differences in particle size, density, shape, surface roughness and also due to interactions between particles (Hogg, 2009). The bed voidage is the main parameter that represents the structure of the bed. Since it is difficult to measure this in large scale reactors, it is either determined through experiments on the laboratory scale or used as an adjustable parameter in mathematical models. However, simple empirical expressions were derived for the bed voidage as a function of the parameters of the particle size distribution such as normalized inter-fractile range, mean or median particle diameter, normalized inter-fractile coefficient of skewness, dispersion of the distribution, and moisture content in case of granules or pellets, and applied for laboratory scale or industrial scale packed bed reactors (Venkataramana et al., 1999; Majumder et al., 2012). Theoretical models for particle packing have also been proposed in the literature (Lee, 1970; Bierwagen and Saunders, 1974; Gan et al., 2004) and were applied to predict voidage or maximum packing fraction as a function of the particle size distribution (Servais et al., 2002; Muralidharan and Runkana, 2009). The DEM was also applied for simulation of packing of spherical particles in a cylinder (Siirii and Yliuruusi, 2007).

Generally the particles are irregular in shape and have a wide size distribution which leads to segregation and non-uniform bed voidage in the axial and radial directions. The particle shape is commonly represented using the sphericity and the flow of particles is modeled by treating the bed of particles as a continuum. More recently, however, the discrete element method (DEM) (Cundall and Strack, 1979) has been applied to simulate flow of particles wherein not only the particles can be treated as individual entities but the particle shape can also be taken into account by creating irregular shaped particles using the sphere as a basic building block.

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2.2 Fluid flow and heat transfer

Intimate mixing of gases and particulate solids is crucial for efficient heat and mass transfer, and reactions between the two phases. In case of rotary drum reactors, the freeboard gases flow mostly over the solids. On the other hand, in case of packed or moving bed reactors, the gases have to pass through a porous bed of particles and also through the particles. Not only the bed voidage but also the particle voidage play an important role here. The Navier-Stokes equations are commonly applied for flow of gases in rotary kilns and in fluidized bed reactors whereas the Ergun equation or the Darcy’s law is employed for flow through porous media.

The heat transfer process in multiphase reactors is complex and involves transfer of heat through radiation, convection and conduction between gases, solids, reactor wall and the environment. The dominant mode of heat transfer between the two phases depends on the nature of flow of solids and gases. Convection is generally the more dominant mechanism for heat transfer between phases and also when the particles are porous in nature, whereas heat transfer occurs mainly through conduction within the solid particles.

Gorog et al. (1981) studied the radiation phenomenon in rotary kilns by evaluation of the fundamental radiative exchange integrals with a modified reflection method using numerical analysis. Barr et al. (1989a, 1989b) conducted experiments with limestone, Ottawa sand and petroleum coke to obtain net rates of heat transfer between solids bed, gas, refractory wall and radial flux for the inside refractory surface as a function of circumferential position. They have also developed a cross-sectional model consisting of a finite difference approximation of conduction in the kiln wall, together with the radiative and convective heat transfer coefficients for setting up the boundary condition. Boateng and Barr (1996) developed a quasi-three-dimensional model for heat transfer between gases and solids in a rotary kiln from a two-dimensional representation of bed transverse plane and one-dimensional conventional plug flow model. The model was shown to predict the temperature distribution in the rolling bed, which tends to be isothermal at higher rotation rates, accurately. Dhanjal et al. (2004) investigated heat transfer in the transverse plane of solids bed in a rotary kiln and developed a model to scale up the experimental results. They used mixing conductivity to simulate the particle mixing effects in the active layer of heat transfer between the two phases which was evaluated by fitting temperature predictions to the experimental data. They have shown that the segregation or particle size has little effect on heat transfer within the bed.

Computational fluid dynamics (CFD) models are applied quite commonly now for simulation of fluid flow, especially because of the availability user-friendly simulation software such as Fluent™, CFX™, and COMSOL™. CFD models have been applied, for instance, for the simulation of fluid flow as well as heat transfer in lime kilns, cement kilns (Mastorakos et al., 1999), for incineration of hazardous waste in a rotary kiln (Yang et al., 2003), for sintering of iron ores (de Castro et al., 2012), for gas flow in induration of iron ore pellets on a moving packed bed reactor (Cross et al., 1990); and for the production of polyethylene in a fluidized bed reactor (Kiašemshahi et al., 2006). Recently, Verma and coworkers (2013, 2014) developed a two-fluid continuum model based on kinetic theory of granular flow to investigate bubble formation in a gas-solid fluidized bed and validated the model with experimental data obtained using ultrafast electron beam X-ray tomography. Since flow of gases and solids influence each other, especially for moving and fluidized bed reactors, efforts have been directed towards coupling the DEM for flow of particles with the CFD model for flow of gases (Tsuji et al., 1992; Tsuji, 1993; Zhou et al., 2008; Yuu et al., 2010; Ueda et al., 2010b). Marias (2003) utilized the process simulation software gPROMSTM for modeling solids processes and coupled it with the CFD tool Fluent™ for modeling fluid flow to simulate a rotary kiln incinerator.

2.3 Evaporation and condensation of moisture

In case of many industrial processes, raw materials are stored in the open and transported through open conveyors. Several materials are hygroscopic in nature and capture moisture during storage and transport, and during rains. Water is sprayed in dry seasons to prevent dusting and it is also used commonly as a liquid binder in many agglomeration processes such as balling and granulation. The kinetics of drying is important because the moisture present in the solids not only delays the onset of reactions between the gases and solids but also causes additional pressure drop, especially in packed bed reactors. Considerable amount of fuel is consumed for evaporation of the moisture from the particles. Similarly, when the product particles are cooled with ambient air, moisture present in the air condenses on the particles and leads to additional pressure drop. The kinetics of drying was represented as a two-step process involving constant rate of drying up to the critical moisture content in the first stage and then falling rate of drying in the second stage (Patisson et al., 1990) in the case of drying of iron ore granules, or as a simple first order rate process (Patisson et al., 2000a) in the case of drying of coal. Although this is an important phenomenon, it has not received as much attention and there is a need to incorporate appropriate models for drying and condensation kinetics in the industrial reactor models for the reasons stated above.
2.4 Reaction kinetics

Both the solid and gas phases are usually multicomponent in nature and unlike controlled experiments in the laboratory, industrial scale reactors involve series as well as parallel reactions both within a phase and between phases. For example, coal gasification and combustion, as well as direct reduction of hematite by carbon monoxide and hydrogen occur simultaneously during the production of sponge iron (Donskoi and McElwain, 2003). The coal gasification reactions are:

\[
\begin{align*}
C (s) + CO_2 (g) & \rightarrow 2 CO (g) \\
C (s) + H_2O (g) & \rightarrow CO (g) + H_2 (g)
\end{align*}
\]

The direct reduction process involves the following series-parallel reactions involving CO and H₂: conversion of hematite (Fe₂O₃) to magnetite (Fe₃O₄):

\[
\begin{align*}
3Fe_2O_3 (s) + CO (g) & \rightarrow 2Fe_3O_4 (s) + CO_2 (g) \\
3Fe_2O_3 (s) + H_2 (g) & \rightarrow 2Fe_3O_4 (s) + H_2O (g)
\end{align*}
\]

Conversion of magnetite to wustite (FeO):

\[
\begin{align*}
Fe_3O_4 (s) + CO (g) & \rightarrow FeO (s) + H_2 (g) \\
Fe_3O_4 (s) + H_2O (g) & \rightarrow 3 FeO (s) + H_2 (g)
\end{align*}
\]

Conversion of wustite (FeO) to metallic iron (Fe):

\[
\begin{align*}
FeO (s) + CO (g) & \rightarrow Fe (s) + CO_2 (g) \\
FeO (s) + H_2 (g) & \rightarrow Fe (s) + H_2O (g)
\end{align*}
\]

The reactions can be exothermic or endothermic in nature. Combustion of gaseous or solid fuels to provide energy for the reaction is a common feature because many gas-solid reactions occur at high temperature. On the other hand, endothermic reactions such as decomposition of CaCO₃ or MgCO₃ to CaO or MgO and CO₂ are common in the case of processes which involve utilization of limestone or dolomite as a fluxing material. The shrinking core model or its variants are commonly used for gas-solid reactions (Omori, 1987; Levenspiel, 1988; Homma et al., 2005). However, since particle porosity or pore size distribution plays an important role in gas-solid reactions, the random pore model has also been employed (Bhatia and Perlmutter, 1980, 1981; Kajitani et al., 2002). Heat and mass transfer and chemical reactions within a single particle or pellet have also been modeled and incorporated into the global reactor model (Kucukada et al., 1994; Patisson et al., 2000a).

2.5 Melting and solidification

In some cases, the high internal temperature in a gas-solid reactor leads to partial or complete melting of the particles depending on the melting point and chemical composition of the materials present. For example, in the case of iron ore sintering and induration, the internal temperature is on the order of 1400 °C which results in partial melting of iron oxides. The molten material solidifies as the temperature decreases during cooling with gases or ambient air before the product is discharged from the reactor. The internal temperature inside a blast furnace can be as high as 2000 °C and results in complete melting of the iron oxides. The melting and solidification phenomena have been incorporated in mathematical models in an approximate manner only (Patisson et al. 1991; Firth and Manuel, 2005). They depend strongly on the temperature and chemical composition of the materials involved. There is a necessity to develop accurate thermodynamic and kinetic models as well as appropriate experimental studies to understand these phenomena.

2.6 Numerical solution and tuning of models

Depending on the nature of the models, that is, linear or nonlinear algebraic equations, and ordinary or partial differential equations, appropriate numerical techniques such as the Gaussian elimination, Newton-Raphson method, Runge-Kutta method, Gear’s method, Finite difference or element or volume method, are commonly employed to solve the equations (Constantinides, 1987; Liu and Bhatia, 2001). For example, the mathematical model for a rotary kiln can be cast as a two-point boundary value problem, with properties of the solids at the feed end and those at the discharge end of the kiln as boundary conditions. The set of equations can be solved either simultaneously or iteratively (Martins et al., 2001; Majumdar et al., 2007). The simultaneous solution is computationally efficient but requires a good initial estimate of the dependent variables. Convergence problems may also arise because a large number of nonlinear equations are solved simultaneously. The iterative technique is more robust and does not require a good initial guess.

In the case of moving packed bed reactors such as traveling grates employed for sintering or induration of iron ores, alternate direction integration schemes are utilized for solving the mass, momentum and energy balance equations (Majumder et al., 2009) from top to bottom or bottom to top for the gas phase, and from left to right for the particles, in line with the motion of the two phases.

As mentioned earlier, mathematical models for industrial gas-solid reactors involve quite a large number of model parameters such as heat transfer coefficients, reaction rate constants, emissivities, etc., which are dependent on the nature of materials and on the reactor operating conditions. It is necessary to tune or customize the model parameters with actual plant measurements so that the
models can be employed in a predictive mode. While it is simple or straightforward to minimize the error between model predictions and plant data for the process variables of importance through a parameter estimation algorithm to obtain numerical values of model parameters, it does not always lead to meaningful values of parameters. This is due to the highly nonlinear nature of the equations involved and due to mutual compensation (Runkana et al., 2005). On the other hand, a staged approach, comprising of testing individual sub-models with representative plant or experimental data to obtain numerical values of the parameters relevant for that phenomenon in the first stage, and then fine-tuning these parameters subsequently while customizing the global model for the reactor is likely to yield realistic and robust model parameters. It is also important to note that the models have to be tested extensively with plant data obtained under different operating conditions and for raw materials having different characteristics. In some cases, it may become necessary to develop look-up tables for model parameters for different raw materials and for widely differing operating conditions so that the end-users can easily choose appropriate numerical values and use the models for predictive simulations and for process optimization.

3. Framework for optimization

The framework for model-based on-line optimization, shown schematically in Fig. 2, is described here. The key components of this framework are a phenomenological model of the process, a statistical or phenomenological or hybrid phenomenological-statistical model for product quality indices and an optimization algorithm. As mentioned in the Introduction, the optimum solution needs to be arrived at by taking cognizance of the characteristics of the raw materials, process operating conditions, equipment and environmental constraints, production targets and product quality indices. The framework described here was implemented for on-line optimization as well as advanced process control of industrial scale gas-solid reactors used for induration of wet iron ore pellets on a traveling grate (Majumder et al., 2012; Cavalcante et al., 2012).

In order to undertake real-time or on-line optimization, it is necessary to integrate the optimization system with the plant’s supervisory control and data acquisition system (SCADA) or the distributed control system (DCS), the laboratory information and management system (LIMS) and if necessary the manufacturing execution system (MES) or the manufacturing operations management (MOM) system so that optimum solutions are obtained using the real-time or near real-time process and laboratory analyses data. Interfacing of the optimization system with the plant automation and information systems is done through an object linking and embedding for process control (OPC) compliant gateway. A real-time database acts as a data buffer for the data transfer between the optimization system and the automation network of the plant.

The phenomenological process model acts as a virtual plant. It accesses data from the real-time database and is used to simulate the process to predict the state variables or the internal conditions of the reactor, for example, temperature, and chemical composition of the gas and solid phases, pressure and velocity of the gas phase, throughout the reactor. This information, along with other data such as raw material characteristics and chemical composition, is used to predict product quality parameters. In other words, the process model and the product quality model act as soft-sensors. The predictions from these models are then utilized by the optimization algorithm along with constraints such as limits on the quality indices, bounds on the manipulated variables, equipment design parameters, and emission control measures, to determine the optimum values of the decision variables. The optimum set points are either downloaded automatically to the SCADA or the DCS or provided as an advisory to the plant engi-
4. Direct reduction of iron ore by coal in a rotary kiln

Direct reduced iron (DRI) or sponge iron is the preferred raw material for secondary steelmaking. India is the largest producer of sponge iron in the world and majority of the sponge iron plants in India utilize non-coking coal as the reductant. Rotary kilns are widely used in India for DRI production because of the ease of operation and control. We have developed DRIKS™, a simulator for DRI production in a rotary kiln. This software tool is based on a fundamental mathematical model for direct reduction of iron ore by coal in a rotary kiln (Runkana et al., 2010b). The mathematical model was customized and validated with data from one of the industrial scale rotary kilns at Tata Sponge Iron Limited (TSIL) and was utilized for process analysis and optimization. Complete details of the mathematical model and the simulation software tool are provided elsewhere (Runkana et al., 2010a, 2010c) and only brief description of the process, mathematical model and results of optimization are presented here.

4.1 Brief process description

The rotary kiln is a refractory lined steel cylinder in which the direct reduction process takes place in a counter-current fashion with iron ore, coal and dolomite fed from the feed end of the kiln while air enters from the discharge end (referred to as primary air). Secondary air is introduced along the kiln length to provide additional oxygen for the chemical reactions occurring inside the kiln and to improve gas mixing. Additionally, air is used to inject coal pneumatically through a blower from the discharge end. Coal acts as the reducing agent as well as the source of energy.

4.2 Iron ore direct reduction model

The mathematical model developed was one-dimensional in nature and it was meant for steady-state simulation of direct reduction of iron ore. The overall kiln model includes appropriate sub-models for kinetics of reactions involving hematite, magnetite, wustite, carbon, oxygen, carbon monoxide and hydrogen, and the flow of solids inside the kiln (see Fig. 3). The model consists of a set of ordinary differential equations, which includes overall material and energy balances for the gas and the solid phases, material balances for individual species in each phase and height of the solids bed along the kiln length. In order to predict gas and solid phase temperature and concentration profiles along the kiln length, the rotary kiln is divided into a number of computational elements over which material and energy balances for the gas and the solid phases and mass balances for the individual species are written. In the direct reduction process various physico-chemical and thermal phenomena occur inside the rotary kiln, which result in transfer of material across phases, either due to phase change, such as evaporation of moisture or volatilization of species present in coal, or due to chemical reactions, such as reduction of various mineral phases or carbon combustion. All such mechanisms were considered in the mass transfer model.

Heat transfer between gases, solids, the kiln refractory wall and the shell occur by different mechanisms, depending on the temperature and the transport medium. The mechanisms of heat transfer that were incorporated in the model are: radiation and convection between gases and the kiln refractory wall; radiation and convection between gases and solids; radiation and conduction between the kiln refractory wall and solids; conduction in the kiln refractory layer; conduction in the kiln shell layer; and convection (natural or forced) and radiation from the kiln shell to the ambient air.

The kinetics of the following reactions or mass transfer processes were incorporated in the model: Drying of particulate matter (solids) like iron ore, coal and dolomite, which happens mainly in the pre-heating zone; Coal devolatilization and gasification; Step-wise reduction of iron ore, from hematite to magnetite, magnetite to wustite and wustite to metallic iron by carbon monoxide and hydrogen, as described in Section 2. The volatile matter in coal was assumed to consist of tar, methane, carbon monoxide, carbon dioxide, hydrogen and chemically-bound water.

The variation of the bed height along the kiln length was related to the total flow rate of solids, kiln internal diameter, angle of inclination of the kiln with the horizontal and the dynamic angle of repose of the solids. This information is used to calculate the angle and degree of filling, which in turn, are used to compute the axial velocity and
mean residence time of the solids inside the kiln. The angle of filling is useful in computing the interfacial area for heat transfer between gases, solids and the kiln wall. Additionally, the model is also capable of predicting the refractory wall and shell temperatures along the kiln length.

### 4.3 Model validation & process optimization

The model was tuned for the operating conditions in one of the rotary kilns at TSIL. The gas and solid temperature profiles, measured using the K-type quick response thermocouples (QRTs) and the metallization (% Fe in DRI), were used for tuning and validation of the model. Typical results for validation of the model are shown in Fig. 4 and Fig. 5 for gas and solid phases, respectively.

The model was used subsequently in a predictive mode to identify optimum conditions of key process variables to increase kiln throughput, to minimize coal consumption and to enhance kiln campaign life. The air profile and the total carbon-to-iron (C/Fe) ratio were found to be critical to achieve the above objectives. The optimum values of these variables were determined through repeated simulations and they were maintained at their set points on a regular basis, which resulted in the reduction of coal consumption by about 55 kg/ton of DRI produced. As a result of this model-based optimization exercise and additional process optimization and control measures undertaken by the plant engineers, the kiln campaign life has also increased beyond 300 days, from approximately 150–180 days for previous kiln campaigns (Runkana et al., 2010b).

![Fig. 3 Schematic of the sponge iron or direct reduced iron kiln model showing sub-models including and inputs and outputs of the model.](image-url)
5. Induration of wet iron ore pellets on a traveling grate

Pelletization is an important process in the iron and steel industry. It is essentially a process for converting raw iron ore fines into hard agglomerates or pellets, which are fed to a blast furnace or a reduction furnace to make hot metal (liquid iron). Pelletization involves two steps,balling of fine particles on a rotating disc into wet pellets, which are then converted into fired pellets on a traveling grate induration furnace (Fig. 6). The quality of fired pellets has a strong influence on the productivity of the reduction furnace and on the quality of hot metal. Hence, maintaining pellet quality within the specifications is critical while optimizing induration to enhance furnace productivity and energy efficiency. One of the key problems encountered by plant personnel is that pellet quality is analyzed off-line through laboratory analyses. Secondly, it is not possible to measure any intrinsic process parameters such as the time-temperature history of pellets because the furnace moves continuously on a traveling grate.

We have developed a software tool, Virtual Indurator™ to address the above problems. The framework for model-based optimization and control of induration of wet iron ore pellets is shown schematically in Fig. 7 (Runkana and Majumder, 2013). This tool consists of a rigorous model of the induration process (Fig. 8) (Majumder et al., 2009) which acts as a soft-sensor for properties of pellets and gases, a set of statistical models derived through data analytics that act as soft-sensors for pellet quality and has an in-built optimization algorithm based on sequential quadratic programming.

5.1 Brief process description

A schematic of the typical straight grate iron ore induration furnace is shown in Fig. 6. The wet pellets are first...
charged onto the moving grate and pass through updraught drying (UDD) and down draught drying (DDD) sections of the reactor for moisture removal from the pellets through evaporation. Subsequently, the temperature of the bed increases slowly in the pre-heating zone (PHZ) using recycled gases. In the firing zone or ignition zone (FZ), the pellet bed is subjected to firing by a set of burners located in the control zones of the ignition hood at the recuperation section of the furnace. As the temperature of the pellet bed increases further, partial melting takes place, leading to fusion within the pellets. After the firing zone, as the pellet bed moves into the after-firing zone (AFZ), the bottom section of the pellet bed is ignited primarily by re-utilizing the enthalpy of the recycled gas stream from first stage cooling (CZ1). After the recuperation section, the pellet bed is cooled in the first (CZ1) and the second stage (CZ2) cooling zones sequentially. To increase the energy efficiency of this energy intensive process, spent gas streams with high enthalpy content are recycled to appropriate stages of the furnace.

5.2 Induration model

The induration process model is based on the fundamental principles of heat and mass transfer between gases and pellets, chemical reaction kinetics for combustion of coke, calcination of limestone (CaCO$_3$), kinetics of drying and condensation of moisture, and partial melting and solidification of the solids. Complete details of the mathematical model can be found elsewhere (Majumder et al., 2009). The model consists of overall energy and material balances for the gas and solid phases, gas pressure drop across the particulate bed, and material balances for individual species in the gas and solid phases. Forced convection is the dominant mode of heat transfer in this reactor.

The energy balance for the gas phase includes terms for convective heat transfer between the gas and solid phases and the heat transfer due to moisture evaporation/condensation. The energy balance for the solid phase includes terms for the convective heat transfer, latent heat of moisture drying or re-condensation and enthalpy due to coke combustion, limestone calcination and pellet melting. The mass balances of the individual components in the gas and the solid phases are formulated appropriately considering moisture evaporation or re-condensation, coke combustion and limestone calcination.

The moisture drying from the pellet was assumed to take place at a temperature midway between solid phase and gas phase temperatures and is represented by two stages, constant and falling rates of drying. During the constant drying rate stage, drying takes place at a constant rate till the moisture reaches the critical moisture level of 5\% (Patisson et al., 1990), after which drying by falling rate stage follows. The coke combustion and limestone calcination reactions within the pellets were assumed to take place according to the shrinking core model (Levenspiel, 1988). The rate expression for kinetics of coke combustion was taken from Wynnyckyj and Batterham (1985) and that for limestone calcination was taken from Clixby and Young (1992).

One of the important inputs to the induration process model is the cold bed voidage which is used for calculating the bed permeability as well as the pressure drop across the particulate bed. This is usually treated as an adjustable parameter in the induration models. In our model, cold bed voidage was calculated using the indices that represent the wet pellet size distribution, namely, mean pellet diameter, normalized inter-fractile range and normalized inter-fractile coefficient of skewness, and moisture content of pellets (Venkataramana et al., 1999).

The quality of fired pellets obtained after induration is represented using parameters such as cold compressive
strength (CCS), tumble index (TI) and abrasion index (AI). These parameters depend not only on the characteristics of the raw materials used but also on the operating conditions during balling and induration, for example, time-temperature history during induration which cannot be measured. Hence, we have developed hybrid phenomenological-statistical models for these parameters, using raw material properties as well as time-temperature history during induration, obtained through simulations using the rigorous induration model (Runkana and Majumder, 2013).

5.3 Model validation & process optimization

The phenomenological induration model was tuned appropriately and validated with sufficiently large sets of process data collected over several months of process operation at two industrial scale pellet plants in India and Brazil (Majumder et al., 2012; Cavalcante et al., 2012), covering diverse operating regimes and raw materials. Model parameters such as heat transfer coefficients, reaction rate constants, etc. were estimated either through trial and error or through nonlinear parameter estimation techniques by minimizing the error between the model predictions and the plant measurements. The accuracy of the phenomenological induration model was verified by comparing the predicted temperature of gases in the wind-boxes below the induration reactor with plant measurements. A typical result for induration process model validation with waste gas temperatures in the wind-boxes is shown in Fig. 9.

After validating the model, process optimization was carried out using an appropriate optimization algorithm. Initially, multi-objective optimization was carried out using a genetic algorithm (Mitra et al., 2009) but since it was computationally intensive, a single objective optimization problem was formulated and solved using the SQP (Majumder et al., 2012). The induration model and the optimization algorithm were tested extensively with several months of process operating data to check numerical convergence, robustness of the model and accuracy of model predictions and optimization solutions obtained. The model-based optimization system was then implemented on-line, as described earlier, by integrating it with the DCS and the LIMS through appropriate software interfaces. Process optimization was then carried out to determine optimum values of manipulated variables to enhance plant production while maintaining product quality within pre-specified constraints. As a result of this on-line optimization exercise, it was possible to improve rate of production by about 3%.

6. Concluding remarks and suggestions for future research

Heterogeneous non-catalytic gas-solid reactors are prevalent in many diverse industries such as chemicals, minerals and metals, polymers and ceramics. Although the fundamentals are well established, application of mathematical models for industrial processes is not very common due to complexity of the multiphase phenomena involved and their interactions and due to the heterogeneous nature of the materials.

Important physico-chemical phenomena that take place during heterogeneous multiphase industrial gas-solid reactors were elucidated in this article. Development of phenomenological process models and their application for on-line process optimization was described along with two industrial case studies on induration of wet iron ore pellets on a traveling grate and direct reduction of iron ore by coal in a rotary kiln. The concepts of hybrid phenomenological-statistical models for product quality and application of rigorous models as soft-sensors for on-line optimization were discussed.

An integrated model coupling charging of the particles, segregation and flow of particles, formation of particulate bed, flow of gases through the particulate bed, heat and mass transfer between the two phases, reactions within and involving the two phases, heat transfer between gases, solids, equipment and the environment, evaporation and condensation of moisture, melting and solidification of the particles has still not been developed. Such a model will be of immense value for optimization of processes such as sintering, induration, direct reduction, calcination, etc.

Formulation of process optimization problems usually incorporate targets for maximizing plant productivity and minimizing energy consumption with constraints on quality or equipment or process variables. There is a need to include constraints on emissions and personnel safety in the problem formulation.
Acknowledgments

The author thanks the management of Tata Consultancy Services for the permission to publish this paper and his colleagues, Sushanta Majumder, Viswanadhan Bandla and Pradeep V. Natekar for their contributions during the implementation of industrial projects. He is also grateful to Prof. E. C. Subbarao, Prof. P. C. Kapur, Prof. Mathai Joseph, Dr. Pradip and Mr. K. Ananth Krishnan for their helpful suggestions and encouragement.

References


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Author’s short biography

Venkataramana Runkana

Venkataramana Runkana is currently a Principal Scientist at Tata Research Development and Design Centre, a division of Tata Consultancy Services. Venkat received B. Tech (Chemical Engineering) from Sri Venkateswara University College of Engineering, Tirupati (India), M.Tech (Chemical Engineering) from Indian Institute of Technology, Kanpur (India) and Ph.D. (Earth and Environmental Engineering) from Columbia University, New York (USA). Venkat has more than 23 years of experience in developing and implementing industrial research projects on process modeling, simulation and optimization, process data analytics, and process development, scale-up and design. His current areas of research are nanomaterials and their applications, colloidal suspensions, drug delivery systems and process data analytics.