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Sulfur Recovery Units: Adaptive Simulation and Model Validation on an Industrial Plant

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The paper is aimed at discussing and fixing issues in providing a generalized approach to the simulation of sulfur recovery units (SRUs). The main goal is to get a simulation that is at the same time (i) reasonably detailed and robust to properly characterize SRUs and (ii) so generalized to provide a tool that is not only specific for the case in study. To achieve point (i), standard libraries belonging to commercial process simulators are coupled to specific heuristic relations coming from the industrial experience for modeling the thermal furnace and the catalytic Claus converters; this allows us to infer with a certain reliability those measures that are often missing or unavailable online in these processes. To achieve point (ii), a series of adaptive parameters are filled in the process simulation by making it more flexible and yet preserving all model details. The most recent techniques and numerical methods, to tune the adaptive simulation parameters, are implemented in Visual C++ and interfaced to PRO/II (by SimSci-Esscor) to obtain a robust parameter estimation solved by means of the BzzMath library. At last, the detailed and tuned adaptive simulation is validated along a period of 2 months on a large-scale SRU (TECHNIP-KTI SpA technology) operating in Italy.

1. Introduction

Data reconciliation has been largely studied, and several approaches and techniques have been proposed in the literature, but from a process and chemical point of view, its applications have been mainly reduced to solve daily/weekly production accounting as well as approximate performance monitoring issues. Moreover, facing the most recent promising approaches such as dynamic reconciliation,[1−3] model-based performance monitoring,[4,5] etc., only the simplest methods have been used to practically tackle the aforementioned problems.

According to Bagajewicz,[6] the reason is that the advanced solutions are still not proven by the field and the commercial cycle of the simplest ones is not yet over. Moreover, some additional reasons should be accounted; in primis, many measures cannot be acquired online by the field (i.e., molar compositions in a thermal furnace, catalyst deactivation, etc.); second, some instrumentation may be too expensive or require long times to get a response. As a consequence, many processes are generally characterized by a lack of process instrumentation leading to an inadequate redundancy and hence making infeasible the data reconciliation.

This lack of information is sometimes so relevant that both field and control room operators cannot know exactly what the current plant condition is. This is the case in those processes that are not directly economically appealing even though their presence is essential for the overall plant. Sulfur recovery units (SRUs) are a typical example since these processes do not directly increase the net present value of the refinery because of the low sulfur market price, but they are necessary to match the more and more stringent environmental regulations. From this perspective, there is the need to develop a detailed model to effectively infer the missing measures and information and hence to make not only feasible but also reliable and robust the data reconciliation.

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This manuscript is specifically aimed at developing a detailed SRU model by means of a field-proven simulation package and applying and validating it on an industrial case study: a large-scale SRU operating in an Italian refinery. Nevertheless, the solution can be easily extended to most of the operating SRUs, thanks to their similar layouts, by inserting a series of adaptive parameters that make the detailed simulation more flexible and not univocally related to the single case in study.

This simulation work is part of a wider research activity aimed at implementing by the field an online solution of rigorous data reconciliation for SRUs. Specifically, the present paper faces the essential step of developing the adaptive simulation and validating it by the field since many authors encountered difficulties in simulating Claus processes and the literature tried to propose some approaches to characterize them without being able to bring them to a generalized framework.[7−9] On this subject, this work has the objective of bridging the gap of scientific literature by fixing the SRU simulation through the combination of theoretical aspects and a long industrial experience in designing and simulating these units. Conversely, activities strictly related to the online application are left to future developments since it directly depends on the success of the adaptive SRU simulation.

The structure of the tool developed here is structured as reported in Figure 1. The raw data coming from the field are preprocessed by opportune robust estimators to purify them by possible gross errors and bad quality points that usually affect the sets of measures. Clean data are used as input for the SRU simulation, which involves some heuristic relations, based on 40 years of industrial experience, inserted for reasonably characterizing thermal furnace and catalytic converters. The simulation is fully integrated with C++ classes belonging to some of the most performing numerical libraries to sequentially solve:

- The detailed SRU simulation.
- The development of an adaptive SRU simulation with the estimation of parameters for the selected industrial case by using the error-in-variable method (EVM), which is based on rigorous data reconciliation theory.[2,10,11]

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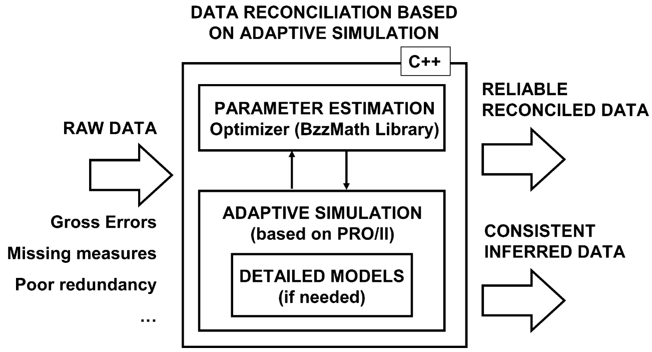

Figure 1. Architecture of the fully integrated approach.

- The tuning of parameters by means of EVM itself to adapt the flexible simulation to the specific case in study.
- The data reconciliation based on the tuned and adaptive SRU simulation.

The proposed approach ensures a reliable inferentiation of missing measures; inferred measures can be used to increase measure redundancy as well as to bridge instrumentation lacks by making feasible the data reconciliation on SRUs. The description of the operating SRU adopted to validate the model is proposed in section 2. Heuristic relations to properly model relevant phenomena and reasonably characterize the complex kinetic mechanisms involved in the thermal furnace as well as in the catalytic reactors are explained in section 3. Theoretical concepts of adaptive simulation are given in section 4. The simulation tuning by means of parameter estimation is performed in section 5. Also, a comparison among different methods for tuning the simulation is provided. Simulation results are discussed in section 6.

2. Process Description

The task of Claus processes is to recover elemental sulfur from hydrogen sulfide and, more generally, from byproduct gases originating from physical and chemical gas and oil treatment units in refineries, natural gas processing, and gasification plants, to quote a few. It consists of a thermal reaction furnace, a waste heat boiler, and a series of catalytic (Claus) reactors and condensers. The overall reaction characterizing the process is as follows:

$$2H_2S + O_2 \rightarrow S_2 + 2H_2O \tag{1}$$

even though complex kinetic mechanisms take place in both the thermal reactor furnace and the catalytic reactors as well.[9] A qualitative layout of a typical Claus process with two catalytic reactors is reported in Figure 2.

In the thermal furnace, one-third of hydrogen sulfide is oxidized to sulfur dioxide using air (or enriched air). Temperatures are usually in the order of 1100−1400 °C. The oxidizing reaction is

$$H_2S + \frac{3}{2}O_2 \rightarrow SO_2 + H_2O \tag{2}$$

which is exothermic and without any thermodynamic restriction.

The two-thirds of unreacted hydrogen sulfide reacts with the sulfur dioxide to produce elemental sulfur through the so-called Claus reaction:

$$2H_2S + SO_2 \leftrightarrows \frac{3}{2}S_2 + 2H_2O \tag{3}$$

which takes place at high temperatures in the thermal furnace with an endothermic contribution or at low temperatures in the catalytic converters with an exothermic contribution.

A secondary but evenly important task is to oxidize ammonia and other impurities usually included in an acid gas (AG) refinery stream. Moreover, some side reactions occurring in the thermal furnace may lower the efficiency in sulfur recovery by generating COS and CS_2. Also, these mechanisms shall be modeled in the process simulation to accurately characterize the SRU as described in the following section.

Off-gas leaving the thermal furnace enters the waste heat boiler where it is quenched to about 300 °C to prevent recombination reactions; then, before entering the catalytic region, the first separation of liquid elemental sulfur is carried out in the first condenser. The hydrogen sulfide conversion goes on in the catalytic region according to the following reaction taking place at low temperature:

$$2H_2S + SO_2 \leftrightarrows \frac{3}{8}S_8 + 2H_2O \tag{4}$$

A condenser is installed downstream each catalytic reactor to condensate and make feasible the separation of the elemental sulfur before entering the next catalytic reactor with the 2-fold advantage of preventing the sulfur condensation into the downstream catalytic reactor and to shift toward the products side the equilibrium of Claus reaction 4.

In addition, COS and CS_2 hydrolysis reactions take place in the catalytic region:

$$COS + H_2O \leftrightarrows CO_2 + H_2S \tag{5}$$

$$CS_2 + 2H_2O \leftrightarrows CO_2 + 2H_2S \tag{6}$$

It is worth underlining that the sulfur recovery of Claus reaction is the most significant one, but if the sulfur recovery requirements are larger than 97.5%, reactions 5 and 6 become relevant, and one has to account for them to accomplish process specification.

One of the most significant indices of the process behavior is the molar ratio H_2S/SO_2 at the tail gas that should be as close as possible to the value of 2 to get the maximum conversion in sulfur (see Figure 3). This index is a controlled variable measured by means of an online analyzer and included in a feedback control where the inlet air feed flow rate is the manipulated variable to close the loop. This measure is particularly sensitive to several process perturbations.

The trend of Figure 3 shows how a small deviation of H_2S/SO_2 ratio in the gas flow rate exiting the thermal furnace takes to large variations in the same ratio at the tail gas and the process simulation shall account for it. This ratio is the most problematic state to be managed in SRU plants, and it usually gives many problems in performing data reconciliation.

The process behaves nonlinearly, and many existing data reconciliation packages, which are not based on rigorous process modeling, easily fail in detecting outliers and in effectively reconciling raw measurements as the process moves away from its conventional operating point. Thus, to adequately simulate the SRU, a deep knowledge of the same process and its peculiarities is essential to develop a reliable toolkit for data reconciliation. The present research activity is the result of the academic−industrial collaboration between Politecnico di Milano and Advanced Process Solutions of Chemprod Srl aimed at combining the experience coming from Technip-KTI SpA, a lead engineering society in designing SRUs, and their design tools with the commercial process simulator PRO/II[12] for

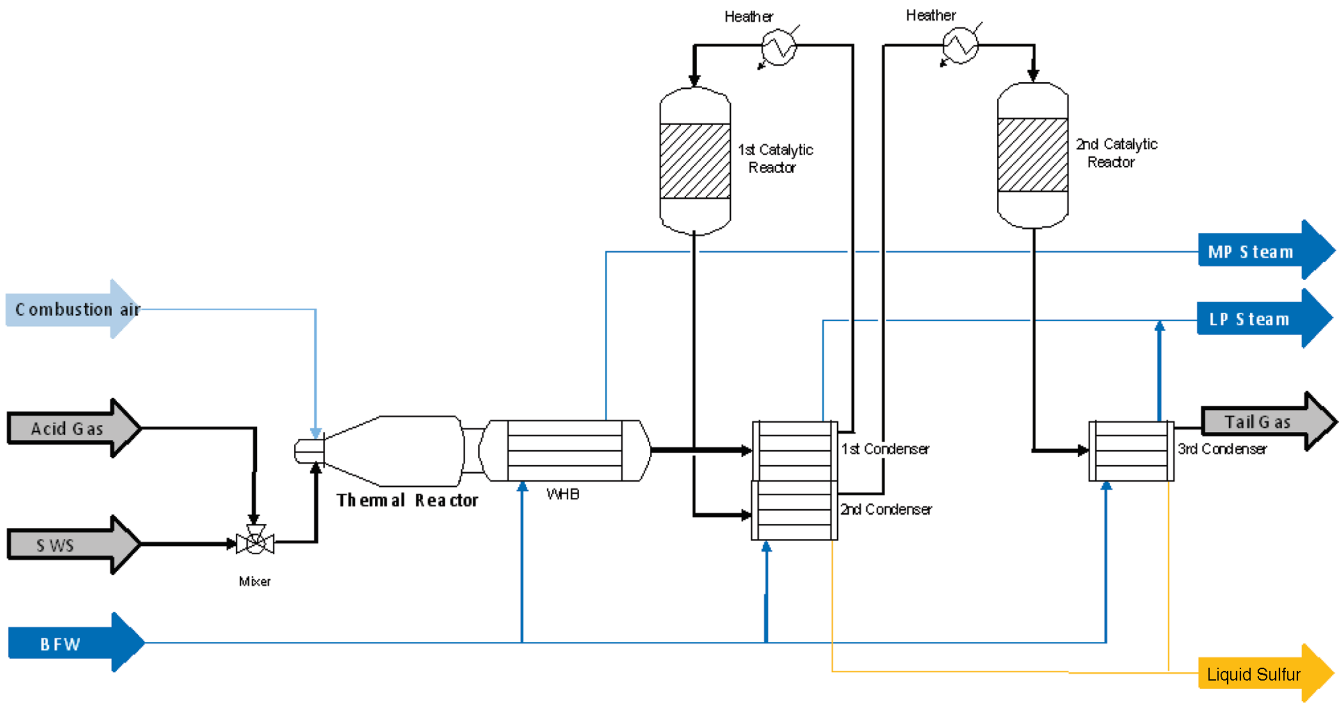

Figure 2. Qualitative layout of the industrial SRU in the study.

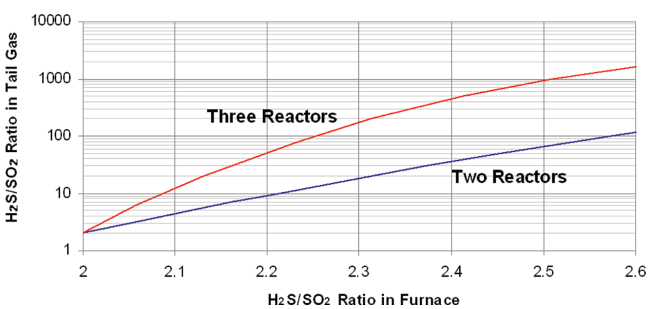

Figure 3. H_2S/SO_2 sensitivity with two and three Claus reactors.

characterizing at the best both thermal and catalytic reactors of the SRU plant as described later. This work is the fundamental basis to achieve a generalized approach to solve the robust and effective data reconciliation tool for these kinds of processes.

3. Adaptive SRU Simulation

The simulation has been developed by means of models belonging to the basic equipment library of PRO/II (about 40 models of reactors, unit operations, heat exchangers, mixers, splitters, etc. were employed). Beyond the essential reactions for H_2S oxidation and for producing elemental sulfur (Claus reaction), the reaction system implemented also includes oxidation of ammonia and light hydrocarbons, formation and hydrolysis of both COS and CS_2, and sulfur equlibria to switch among S_2, S_6, and S_8.

The philosophy adopted is to distinguish process lines from the steam generation network. The process line is modeled as a series of Gibbs reactors, which allow consideration of thermodynamic equilibria among the sulfur species in accordance with the operating temperature of condensers, preheaters, and waste heat boiler. Conversely, models to simulate the steam generation network account even for energy balances. To improve the accuracy, blowdown of steam generators has been considered, too.

Oxidation mechanisms taking place in the thermal furnace are modeled as full conversion reactions. On the other hand, the Claus reaction at high temperature is characterized by a partial conversion according to the hydrogen sulfide fraction of the feed flow rate. The heuristic relation describing the H_2S conversion at high temperature $\xi_{\text{Claus high }T}$ is as follows:

$$\xi_{\text{Claus high }T} = \frac{4}{3}\frac{\dot{n}_{S_2}^{OUT} - \dot{n}_{S_2}^{IN}}{\dot{n}_{H_2S}^{IN}} \quad (7)$$

The molar flow rate of the reacted H_2S is here referred to as the molar flow rate of S_2 produced ($\dot{n}_{S_2}^{OUT} - \dot{n}_{S_2}^{IN}$) multiplied by the H_2S/S_2 stoichiometric ratio as it appears in reaction 3. The $\dot{n}_{S_2}^{OUT}$ can be then calculated as follows:

$$\dot{n}_{S_2}^{OUT} = \left[0.2040\left(x_{H_2S}^{AG}\right)^4 - 0.6831\left(x_{H_2S}^{AG}\right)^3 - 0.2293\left(x_{H_2S}^{AG}\right)^2 + 1.6962\left(x_{H_2S}^{AG}\right) - 0.2939\right]\dot{n}_{S_{2}EQ}^{AG} \quad (8)$$

where $x_{H_2S}^{AG}$ is the molar fraction of H_2S in the AG flow entering the thermal furnace, $\dot{n}_{S_{2}EQ}^{AG}$ is the total molar flow rate of equivalent (EQ) elemental sulfur, $\dot{n}_{S_2}^{OUT}$ is the molar flow rate of elemental sulfur present in the furnace outlet stream, and $\dot{n}_{S_2}^{IN}$ and $\dot{n}_{H_2S}^{IN}$ are the elemental sulfur and hydrogen sulfide molar flow rates, respectively, in the inlet stream. Coefficients of the fourth-order polynomial form (eq 8) are not directly involved in the operations of tuning (see the next section), and they are considered as constants in this specific case. Nevertheless, each of these parameters could be whenever considered as an adaptive parameter in the most general case and, hence, as a degree of freedom in the parameter estimation problem.

COS and CS_2 formation mechanisms are satisfactorily described by reverse reactions 5 and 6, respectively, by defining a hydrolysis effect. These mechanisms take place even at the first Claus converter where both COS and CS_2 conversions become a significant tuning parameter as described hereinafter.

Assuming that the inlet fraction of COS and CS_2 is practically null, their formation in the thermal furnace can be reasonably defined by the following heuristic relations:

$$\xi_{\mathrm{cos}} = \frac{\left[\theta_1 \frac{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}} \times \dot{n}^{\mathrm{AG}}_{\mathrm{H_2S}}}{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}} + \dot{n}^{\mathrm{AG}}_{\mathrm{H_2S}}} + 0.23 \frac{\dot{n}^{\mathrm{AG}}_{\mathrm{CEQ}} \times \dot{n}^{\mathrm{AG}}_{\mathrm{H_2^{EQ}}}}{\dot{n}^{\mathrm{AG}}_{\mathrm{CEQ}} + \dot{n}^{\mathrm{AG}}_{\mathrm{H_2^{EQ}}}}\right]}{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}}} \quad (9)$$

$$\xi_{\mathrm{CS_2}} = \frac{\left[\theta_2 \times \frac{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}} \times \dot{n}^{\mathrm{AG}}_{\mathrm{H_2S}}}{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}} + \dot{n}^{\mathrm{AG}}_{\mathrm{H_2S}}} + 4.02 \times 10^{-3} \times \frac{\dot{n}^{\mathrm{AG}}_{\mathrm{CEQ}} \times \dot{n}^{\mathrm{AG}}_{\mathrm{H_2^{EQ}}}}{\dot{n}^{\mathrm{AG}}_{\mathrm{CEQ}} + \dot{n}^{\mathrm{AG}}_{\mathrm{H_2^{EQ}}}}\right]}{\dot{n}^{\mathrm{AG}}_{\mathrm{CO_2}}} \quad (10)$$

As they are yet accounted aside at the first term of the numerator, it is worth saying that the contributions of carbon dioxide and hydrogen sulfide are not included in the evaluations of the total molar flow rates of EQ carbon $\dot{n}^{\mathrm{AG}}_{\mathrm{CEQ}}$ and of EQ hydrogen $\dot{n}^{\mathrm{AG}}_{\mathrm{H_2^{EQ}}}$, respectively. Contrary to the other coefficients, whose values were aprioristically fixed, the parameter θ_1 is significant to match the COS and the parameter θ_2 to match CS_2 at the tail gas.

The other relevant parameters to tune the SRU simulation and their corresponding influences on process variables are resumed hereafter:

- $\theta_3,\theta_4 \rightarrow$ COS and CS_2 hydrolysis reactions at the first Claus reactor. The tail gas COS and CS_2 compositions are directly influenced even by these parameters. In addition, even the H_2S and SO_2 compositions are slightly modified.
- $\theta_5 \rightarrow \Delta T$ approach on Claus converters. The larger ΔT is, the larger the H_2S and SO_2 compositions are at tail gas. Conversely, the original H_2S/SO_2 ratio is preserved. Actually, ΔT approach is a device to account for the equilibrium nonideality of catalytic reactors.
- $\theta_6 \rightarrow$ thermal loss coefficient at the waste heat boiler. Heat losses are fixed to use them for matching the steam generated in these units without influencing any other process variable. Besides the possibility to match energy balances, thermal losses provide a more coherent picture of waste heat boiler conditions, with the consequent result of adjusting compositions and equilibria ($Q_{\mathrm{exchanged}} = \theta_6 \cdot Q^{\mathrm{WHB}}_{\mathrm{generated}}$).
- $\theta_7,\theta_8 \rightarrow$ heat losses for first and second Claus reactors. These parameters allow matching of the outlet temperature and the molar flow rate of H_2S and SO_2 at the tail gas, according to ΔT approach parameter.

The introduction of adaptive parameters is crucial for two reasons:

1. It is important to have a flexible SRU simulation able to properly follow the process evolutions on the long-term by accounting for the catalyst deactivation, heat exchanger fouling, cleanliness factors, etc. This is easily achieved by retuning parameters with a certain frequency (e.g., monthly).
2. The same flexibility should be extended from the single plant to all plants characterized by similar layouts so to make the detailed (hence specific for the single application) approach generalized.

The sensitivity of main process variables with respect to the aforementioned parameters is quantitatively reported in Table 1. The highest degree of sparsity of the sensitivity matrix points out the fact that each process variable is significantly influenced by one or two parameters only and the contributions of all remaining parameters are practically negligible.

Thus, the adaptive parameter estimation could be probably simplified by estimating one parameter at a time as they are practically uncorrelated. The following paragraph shall consider even this kind of simplified approach by comparing it to the most comprehensive ones proposed in the literature.

4. Theoretical Concepts of Adaptive Simulation

The simulation tuning, as stated before, has been accomplished by means of the so-called EVM.[13−16] This method not only uses the data reconciliation during its solution phase but also has a mathematical formulation similar to data reconciliation itself.

The problem of data reconciliation (paragraph 2.1) could be reduced to a constrained least-squares minimization:

$$\begin{cases} \min\limits_{\mathbf{x}} \Phi = (\mathbf{m} - \mathbf{x})^{\mathrm{T}} \mathbf{Q}^{-1} (\mathbf{m} - \mathbf{x}) \\ s.t.: \\ g(\mathbf{x}) = 0 \\ h(\mathbf{x}) \le 0 \end{cases} \quad \text{data reconciliation} \quad (11)$$

where Φ is the objective function; $\mathbf{x}$ and $\mathbf{m}$ are the vectors of reconciled and measured values, respectively; $\mathbf{Q}$ is the variance and covariance matrix; and $g(\mathbf{x}) = 0$ and $h(\mathbf{x}) \le 0$ are equality and inequality constraints, to which the minimization problem is subject.

The crucial difference between EVM and data reconciliation is in the numerical problem dimensions since more steady-state conditions (SSCs) are considered as follows:

$$\begin{cases} \min\limits_{\mathbf{x}_i,\boldsymbol{\theta}} \Phi = \sum\limits_{i=1}^{\mathrm{SSC}} (\mathbf{m}_i - \mathbf{x}_i)^{\mathrm{T}} \mathbf{Q}^{-1} (\mathbf{m}_i - \mathbf{x}_i) \\ s.t.: \\ f(\mathbf{x}_i, \boldsymbol{\theta}) = 0 \end{cases} \quad \text{EVM} \quad (12)$$

Moreover, degrees of freedom of the minimization problem are not only the unknowns $\mathbf{x}$, but they even include the vector of parameters $\boldsymbol{\theta}$. The variance and covariance matrix $\mathbf{Q}$ does not necessarily correspond to one of the formulation (formulation 11) as, in this case, variances are evaluated by accounting for the selected set of SSC.

4.1. Data Reconciliation. Formulation 11 involves a series of data reconciliation problems in accordance with the nature of constraints, from the first applications involving linear constraints only to the most recent methodologies that include both nonlinear and differential constraints to account for complex dynamic behaviors.

Without entering into details of mathematical formulation that have already been proposed elsewhere,[4,17−19] only a brief remainder is proposed hereinafter. The linear technique is properly employed in facilities and utilities for steam generation, where the only component is water (other components are negligible). Hence, these processes do not require any in-line analyzer to measure molar compositions. Again, they are often characterized by small redundancies in measures, especially for the reduced interest to place some instrumentation on all condensed water lines. By chance, the overall mass and energy balances are enough to characterize the process.

Contrary to steam generation utilities and facilities, when the process flow rates are multicomponents, the reconciliation problem is to be extended from the linear case at least to the bilinear one. Actually, it is necessary to reconcile not only the overall material flow rate but also the mass component rate. It unavoidably requires some in-line analyzers, besides the flow measures. From a computational point of view, it is useful to keep the problem linear when possible as shown by Crowe and co-workers.[20,21] With respect to the aforementioned techniques, the nonlinear data reconciliation has fast acquired interest in

Table 1. Parametric Sensitivity against Process Variables ("S" Stands for Strong Sensitivity, "W" Stands for Weak Sensitivity, and Empty Cells Mean No Correlations)

	parameters							
	θ_1	θ_2	θ_3	θ_4	θ_5	θ_6	θ_7	θ_8
WHB steam flow rate						S		
1st converter outlet temperature					S		S	
2nd converter outlet temperature								S
H_2S composition at the tail gas	W	W	W	W	S		W	S
SO_2 composition at the tail gas	W	W	W	W	S		W	S
total sulfur								S

process industry since it gives the opportunity to reconcile temperature and pressure measurements with a very successful and robust practice.[6]

The fact that some field-proven process simulators are largely spread in process industries (PRO/II by SimSci-Esscor, Invensys; UNISIM by Honeywell; ASPEN HYSYS by Aspen Technology) makes them particularly useful to carry out the nonlinear data reconciliation. Unfortunately, these commercial packages adopt a sequential solver, whereas Lid and Skogestad[22] underlined that optimization problems are better solved by simultaneous approaches provided by ad hoc tools like gPROMS (PSE), ASCEND (Carnegie Mellon University), DATACON (SimSci-Esscor, Invensys), etc.

Nevertheless, the larger diffusion in the process industry of commercial process simulators rather than the specific aforementioned data reconciliation tools pushed us toward the possibility of integrating them as black-box in solving the reconciliation problem. Such a solution may take to a series of benefits:

- The mathematical model can cover each level of detail according to the model libraries proposed by the commercial simulator. The selection of degree of detail should be a good compromise between the process characterization and the computational effort.
- Some consolidated solutions, especially dictated by the practical experience and already implemented in process simulators, could be successfully involved in the solution of data reconciliation problems.
- When using a commercial simulator that is even adopted for process design, the data reconciliation may have a feedback on both the instrumentation and the same process design (plant debottlenecking, revamping, etc.).
- At last, engineering societies and production sites usually have the licenses of process simulators (reduced license duties).

The current frontier of research activities and development deals with dynamic data reconciliation where constraints are differential and differential−algebraic systems.[1,18,23] However, it is still too computationally intensive for industrial applications, and some issues concerning robustness and reliability may arise while applying it. Therefore, its application is confined to the single process unit, whereas the overall plant is still subject to the steady approach. Another limitation is represented by the fact that steady-state reconciliation is not commercially over, especially in its nonlinear case.

4.2. Gross Error (Outlier) Detection. Rather than developing dynamic data reconciliation solutions, several scientific areas mainly focused their attention on the detection of gross errors affecting raw data coming from the field during the last years. In all scientific and industrial areas involving experimentations and plant data, there is the need of identifying gross error existence, location, size, and type, and many statistical tests and methods were developed to solve this hard problem.

One reason that makes the gross error detection a multifaceted problem is the fact that gross errors have no general definition that is well-accepted by everyone, and this often takes to equivocations and errors in speaking about them. The present paper assumes the recent definition proposed in the literature,[24] where whatever gross error is ever joined to the specific model that one is analyzing.

As already demonstrated by Rousseeuw,[25] the method of least sum of squares that was traditionally adopted to point out gross errors is inadequate, and its use should be avoided as both the arithmetic meaning and the standard deviation involved are nonrobust estimators, and they are biased even though there is only a single gross error.

It is not a coincidence that many people proposed robust methods[25−31] to identify them. Notwithstanding, robust methods are not yet widely used, and several reasons may help to explain their unpopularity.[32,33] A possible reason is that computation of robust estimates is much more computationally intensive than least sum of squares estimation; in recent years, however, this objection has become less relevant as computing power has greatly increased. However, the most important reason for the current poor appeal of robust methods in looking for gross errors is that many statisticians erroneously believe that classical methods are sufficiently robust; hence, no more robust alternatives seem to be required.

Alternatives that are more robust than the least sum of squares are the least median of squares,[25,29] the least trimmed sum of squares,[30] and the least clever sum of squares.[24,26] This latter method not only allows one to effectively detect gross errors by self-regulating its own robustness according to the same set of plant data that one is processing but also allows one to preserve the higher efficiency of traditional estimators.

4.3. Instrumentation Upgrade. The availability of instrumentation and measures is usually poor, and data reconciliation cannot be fully and effectively performed. Some rigorous approaches have been used to face this problem by finding the optimal solution involving costs of the instrumentation and benefits deriving from an effective data reconciliation as well.[34−37] As a result, while designing a plant, instrumentation costs are minimized by accounting for the threshold of having a reasonable amount and disposition of process measures to get all benefits of reliable data reconciliation. Conversely, if the plant is already operating and many measures are missing, only an inference of some critical measures may support data reconciliation, making it feasible.

5. Estimation of Adaptive Parameters

Having defined mathematical details of SRU simulation and its parametric sensitivity, it is possible to tune it by estimating parameters $\boldsymbol{\theta}$. The tuning allows adaptation of the generalized simulation to the specific case in study, which is a large-scale operating plant placed in Italy and based on TECHNIP-KTI SpA know-how.

Table 2. Summary of Plant Measures Acquired by the Field, Dependent/Independent Variables for Adaptive Simulation, and Terms Involved in the Objective Function[a]

measures		degrees of freedom		
H_2O		computed		OBJ FUNCTION
CO_2	measured	independent	measured	OBJ FUNCTION
H_2S	measured	independent	measured	OBJ FUNCTION
H_2O		computed		OBJ FUNCTION
H_2S	measured	independent	measured	OBJ FUNCTION
NH_3	measured	independent	measured	OBJ FUNCTION
total H_2S		calculated		calculated
	simul input	simulated	recon input	minimization
AG flow	measured	independent	measured	OBJ FUNCTION
AG temperature	measured	independent	measured	OBJ FUNCTION
SWS flow	measured	independent	measured	OBJ FUNCTION
SWS temperature	measured	independent	measured	OBJ FUNCTION
combustion air flow	measured (guess)	dependent	measured	OBJ FUNCTION
combustion air temperature	measured	independent	measured	OBJ FUNCTION
furnace temperature		dependent		OBJ FUNCTION
WHB temperature	measured	independent	measured	OBJ FUNCTION
tout first condenser	measured	independent	measured	OBJ FUNCTION
tin first Claus	measured	independent	measured	OBJ FUNCTION
tout first Claus	measured	independent	measured	OBJ FUNCTION
tout second condenser		dependent		OBJ FUNCTION
tin second Claus	measured	independent	measured	OBJ FUNCTION
tout second Claus		dependent		OBJ FUNCTION
steam generated at the WHB		dependent		OBJ FUNCTION
H_2S at the tail gas		dependent		OBJ FUNCTION
SO_2 at the tail gas		dependent		OBJ FUNCTION
H_2S/SO_2 ratio		calculated		OBJ FUNCTION
recovered sulfur		calculated		calculated
sulfur conversion		calculated		calculated

[a] Note that the minimization objective function includes both measured and inferred values. Also, the combustion air flow measure is used as an initial guess in process simulation, as it is a dependent variable.

As stated before in paragraph 4, the EVM formulation is close to data reconciliation, but the problem dimension is somehow enlarged. Actually, the resulting minimization problem of data reconciliation has $N_{\mathbf{X}}$ degrees of freedom, where $N_{\mathbf{X}}$ is the amount of reconciled variables; the EVM approach has $N_{\mathbf{X}} \times \mathrm{SSC} + N_{\boldsymbol{\theta}}$ degrees of freedom, where $N_{\boldsymbol{\theta}}$ is the amount of adaptive simulation parameters.

According to the sensitivity analysis of Table 1, one could think to simplify the mathematical formulation of EVM (formulation 12) as follows:

$$\begin{cases} \min\limits_{\mathbf{x}_i,\theta_j} \Phi = \sum\limits_{i=1}^{\mathrm{SSC}} (\mathbf{m}_i - \mathbf{x}_i)^{\mathrm{T}} \mathbf{Q}^{-1} (\mathbf{m}_i - \mathbf{x}_i) \\ s.t.: \\ f(\mathbf{x}_i, \theta_j) = 0 \end{cases} \quad \forall \theta_j \text{ with } j = 1, ..., 8 \tag{13}$$

as adaptive parameters are practically uncorrelated each other. By doing so, the computational effort could be reduced. We will call this approach simplified EVM (S-EVM) in the following.

The problem dimension is enlarged against the data reconciliation of formulation 11. The S-EVM formulation of formulation 13 allows reduction of the overall minimization problem into $N_{\boldsymbol{\theta}}$ minimizations with a dimension equal to $N_{\mathbf{X}} \times \mathrm{SSC} + 1$. Thus, if it could be properly applied (because each parameter practically influences one process variable only, without having any other strong correlations), the S-EVM method could be numerically useful when the amount of reconciled variables and the set of SSC are both small against the number of adaptive simulation parameters.

Even though it is not so significant, a reduction in computational time is granted, especially when the number of adaptive parameters is relatively large if compared to the summation of unknowns $\mathbf{x}$ of all selected SSC. Unfortunately, the contribution of $N_{\mathbf{X}} \times \mathrm{SSC}$ is usually larger than $N_{\boldsymbol{\theta}}$: If we consider that in the specific industrial case here examined $N_{\mathbf{X}} = 24$, $\mathrm{SSC} = 5$, and $N_{\boldsymbol{\theta}} = 8$, the resulting NLP problem dimension only slightly decreases by switching from EVM (128 degrees of freedom for the NLP) to S-EVM (121 degrees of freedom for the NLP) (Table 2).

Notwithstanding, by tuning one parameter at a time (therefore by using the S-EVM), the degree of redundancy increases by giving the possibility to operate one of the following further improvements:

- It is possible to use the same number of SSC to get an increased degree of redundancy, and the approach is more robust in detecting possible outliers.
- It is possible to discard some SSC to further reduce the minimization problem size but only if the robustness is reasonably preserved so to identify possible gross errors.

An additional simplification that significantly reduces the problem dimension may be solving $N_{\boldsymbol{\theta}}$ one-dimensional minimizations where the only degree of freedom is the parameter θ_i, but a consequent loss in robustness and therefore in the possibility of detecting gross errors is assured. For the sake of completeness, even this approach is considered in the following comparison of parameter estimation methods, and it will be named one-dimensional regression (in the following, ODR). The resulting nonlinear constrained multidimensional minimization problem of EVM is solved by adopting the object-oriented VISUAL C++ programming language and the robust optimizers based on OPTNOV method[38,39] that belongs to the freely downloadable BzzMath library.[26,40−42]

It is worth remarking that the estimation of adaptive parameters does not require any online retuning, but such an operation is performed either once, just to adapt the generalized

Table 3. Comparing Approaches for the Estimation of Adaptive Simulation Parameters[a]

	parameter description	ODR	S-EVM	EVM
θ_1	COS conversion coefficient in the thermal furnace	0.0387	0.0385	0.0240
θ_2	conversion of COS hydrolysis reaction	0.0301	0.0301	0.0301
θ_3	CS_2 conversion coefficient in the thermal furnace	0.95	0.95	0.95
θ_4	conversion of CS_2 hydrolysis reaction	0.74	0.75	0.81
θ_5	ΔT approach on Claus converters (°C)	− 14.5	−14.5	−14.2
θ_6	thermal loss coefficient at the WHB (%)	6.5	2.1	5.1
θ_7	heat losses for first Claus reactor (kcal/h)	-4.2×10^4	-4.17×10^4	-3.7×10^4
θ_8	heat losses for second Claus reactor (kcal/h)	-2.1×10^4	-2.3×10^4	-2.2×10^4

[a] ODR is the one-dimensional regression solved accounting for a single SSC; S-EVM is the simplified EVM accounting for one adaptive parameter at a time; EVM is the overall method described in the scientific literature.

Figure 4. AG flow rate entering the SRU during the 2 month period analyzed. Circles represent the five test runs adopted to tune the adaptive simulation parameters to the specific case in study; squares represent the three test runs adopted as validation cases for the adapted simulation. It is worth underlining that data coming from the plant are an hourly average value that ensures that the plant could be considered at steady conditions.

simulation to the plant, or with very low frequency to account for the plant evolution on the long-term (catalyst deactivation, fouling factors, etc.). Consequently, an accurate estimation by means of robust algorithms is preferred to other more computationally performing solutions.

Table 3 proposes a comparison among the aforementioned methods for adaptive parameter estimation. The one-dimensional regression approach usually fails when a single gross error affects the industrial data set because of its limited redundancy. Moreover, it does not equally distribute the stochastic errors on the measures but only on the parameters, making their estimation not so reliable. Even the S-EVM seems not to be so reliable in estimating parameters, as it is unable to simultaneously account for all contributions coming from more parameters when they influence the same process variables. Conversely, the EVM is surely preferable for its robustness, as

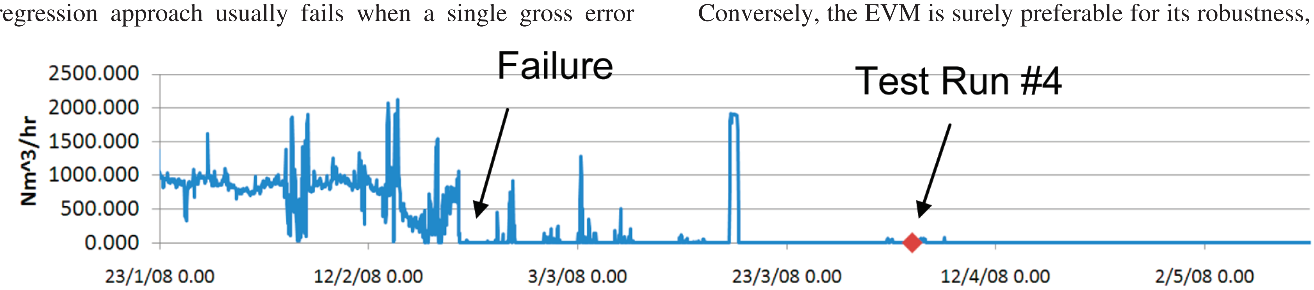

Figure 5. Fault measure on the SWS line and corresponding gross error identification within the data of test run #4.

it is the only one able to account for each rectangular contribution in the parametric sensitivity.

Therefore, \marginally, some parameters are related each other in the parametric sensitivity and the EVM method is the only one able to account for these interactions. So, there is also the possibility to introduce another approach that is intermediate between S-EVM and EVM: It is a reduced EVM (R-EVM) that estimates all together those parameters (subsets of parameters) that present any kind of interaction in the parametric sensitivity against process variables (either strong or weak correlations), by bringing just to a reduced NLP as all remaining parameters are singularly estimated by adopting the S-EVM. Only the total amount of all interacting parameters must be considered as additional degrees of freedom and the maximum dimension of the resulting subset of minimization problems is $N_{\mathbf{X}} \times \mathrm{SSC} + \max_{i=1,\dots,\mathrm{S}} N_{\boldsymbol{\theta}_{\mathrm{C}},i}\}$, with $\max_{i=1,\dots,\mathrm{CS}} N_{\boldsymbol{\theta}_{\mathrm{C}},i}\}$ the maximum of the amount of correlated parameters $N_{\boldsymbol{\theta}_{\mathrm{C}}}$ included in the i-th correlation set CS. Parameter estimations obtained by means of R-EVM methods are very similar to the ones provided by EVM only for the subsets of interacting parameters, whereas all of the other parameters present the same values obtained by S-EVM. Facing a slight reduction in the computational effort, the R-EVM still presents the same issues of poor robustness already discussed for the S-EVM.

It is worth underlining that the model parameter estimation is usually a very hard problem as it may involves a series of additional issues that were intentionally neglected here such as heteroscedasticity condition, multicollinearity, masking and swamping effects, and so on. For the sake of conciseness, we remind the reader to specific scientific contributions[24,26,27,43,44] for detailed discussions on these problems.

6. Simulation Results

After modeling and tuning the process by means of EVM, a wide operational range of conditions of the industrial case in study along a period of 2 months has been considered to validate the simulation. The plant data concerning the AG flow rate entering the plant is reported in Figure 4. The same figure provides:

- Dates of the five test runs (circles) adopted to set up EVM to tune the adaptive simulation parameters and hence to adapt the SRU simulation to the specific case in study.
- Dates of some test runs (squares) adopted as validation cases for the tuned simulation.

These two sets of test runs are intentionally far from each other (see Figure 4) and at different operating conditions, as SRU is processing about 3000 kg/h at the former conditions, whereas it processes less than 2500 kg/h of AG flow rate at the latter ones to validate the flexibility of the approach against possible load changes. Note that some residuals are zero since such measured values are independent variables for the adaptive simulation.

The robustness of the approach can be seen by examining results concerning the eight test runs from Tables 4 to 12. Specifically, three test runs (Tables 4−6) do not show any significant deviation between measured and simulated values as the relative residuals are small. Only the combustion air is a bit higher for the simulation against its corresponding measure, but the gap is in the order of 2−4%.

Conversely, test run #4 of Table 7 shows a relevant deviation between some measures and their corresponding simulated values. The simulation provides a combustion air flow that is smaller than the measured one: about −800 kg/h with a relative deviation in the order of −15%. In addition, according to this simulated value of combustion air, the simulation provides results that are significantly far from data coming from the field: smaller furnace temperature (about −40 °C and −3% of relative deviation); smaller steam generated in the waste heat boiler (−900 kg/h and −15% of relative deviation); and contrary to all of the aforementioned cases, a final H_2S/SO_2 ratio that is slightly different from the measured one. If the single measurement of both H_2S and SO_2 concentrations is obtained by instrumentations often error prone and not widely spread in the process industry, the only way to motivate the relevant deviations between some other measures and the corresponding result of the adaptive simulation is to admit the presence of at least

Table 4. Measured and Simulated Values for Test Run #1 Adopted in Implementing the EVM for Tuning the Adaptive Simulation

measures	test run #1: February 12, 2008, 9 am		
H_2O	0.0875 mol. fract.		
CO_2	0.0884 mol. fract.	AG	
H_2S	0.8241 mol. fract.		
H_2O	0.3191 mol. fract.		
H_2S	0.2873 mol. fract.	SWS	
NH_3	0.3963 mol. fract.		
total H_2S	51 t/day	51 t/day	
	measured	simulated	residuals
AG flow	2180.88 kg/h	2180.88 kg/h	0.00
AG temperature	55.10 °C	55.10 °C	0.00
SWS flow	779.90 kg/h	779.90 kg/h	0.00
SWS temperature	84.67 °C	84.67 °C	0.00
combustion air flow	5205.34 kg/h	5384.75 kg/h	3.45
combustion air temperature	228.73 °C	228.73 °C	0.00
furnace temperature	1394.59 °C	1391.50 °C	−0.22
WHB temperature	305.29 °C	305.29 °C	0.00
tout first condenser	166.79 °C	166.79 °C	0.00
tin first Claus	229.25 °C	229.25 °C	0.00
tout first Claus	291.53 °C	292.07 °C	0.19
tout second condenser	165.53 °C	165.53 °C	0.00
tin second Claus	215.07 °C	215.07 °C	0.00
tout second Claus	221.93 °C	222.27 °C	0.15
steam generated at the WHB	5210.92 kg/h	5127.34 kg/h	−1.60
H_2S at the tail gas	5063 ppmv	5049.0 ppmv	−0.27
SO_2 at the tail gas	1679 ppmv	1678.0 ppmv	−0.03
H_2S/SO_2 ratio	3.02	3.01	−0.23
recovered sulfur		47.25 t/day	
sulfur conversion		92.9749	

Table 5. Measured and Simulated Values for Test Run #2 Adopted in Implementing the EVM for Tuning the Adaptive Simulation

measures	test run #2: February 12, 2008, 1 pm		
H_2O	0.0862 fraz mol		
CO_2	0.0882 fraz mol	AG	
H_2S	0.8255 fraz mol		
H_2O	0.2798 fraz mol		
H_2S	0.3039 fraz mol	SWS	
NH_3	0.4163 fraz mol		
total H_2S	51 t/day	51 t/day	
	measured	simulated	residuals
AG flow	2160.57 kg/h	2160.57 kg/h	0.00
AG temperature	55.22 °C	55.22 °C	0.00
SWS flow	806.78 kg/h	806.78 kg/h	0.00
SWS temperature	84.76 °C	84.76 °C	0.00
combustion air flow	5288.66 kg/h	5521.30 kg/h	4.40
combustion air temperature	229.02 °C	229.02 °C	0.00
furnace temperature	1392.17 °C	1405.62 °C	0.97
WHB temperature	305.48 °C	305.48 °C	0.00
tout first condenser	166.75 °C	166.75 °C	0.00
tin first Claus	229.50 °C	229.50 °C	0.00
tout first Claus	291.85 °C	291.55 °C	−0.10
tout second condenser	165.42 °C	165.42 °C	0.00
tin second Claus	215.75 °C	215.75 °C	0.00
tout second Claus	222.48 °C	222.66 °C	0.08
steam generated at the WHB	5289.65 kg/h	5279.76 kg/h	−0.19
H_2S at the tail gas	5347 ppmv	5312 ppmv	−0.65
SO_2 at the tail gas	1509 ppmv	1502 ppmv	−0.46
H_2S/SO_2 ratio	3.54	3.54	−0.19
recovered sulfur		47.59296 t/day	
sulfur conversion		92.93127	

Table 6. Measured and Simulated Values for Test Run #3 Adopted in Implementing the EVM for Tuning the Adaptive Simulation

measures	test run #3: February 13, 2008, 1 am		
H_2O	0.0824 fraz mol		
CO_2	0.0846 fraz mol	AG	
H_2S	0.8329 fraz mol		
H_2O	0.2967 fraz mol		
H_2S	0.2968 fraz mol	SWS	
NH_3	0.4065 fraz mol		
total H_2S	53 t/day	53 t/day	
	measured	simulated	residuals
AG flow	2304.87 kg/h	2304.87 kg/h	0.00
AG temperature	54.86 °C	54.86 °C	0.00
SWS flow	712.92 kg/h	712.92 kg/h	0.00
SWS temperature	85.45 °C	85.45 °C	0.00
combustion air flow	5388.64 kg/h	5536.09 kg/h	2.74
combustion air temperature	217.34 °C	217.34 °C	0.00
furnace temperature	1395.64 °C	1391.72 °C	−0.28
WHB temperature	307.48 °C	307.48 °C	0.00
tout first condenser	166.76 °C	166.76 °C	0.00
tin first Claus	229.26 °C	229.26 °C	0.00
tout first Claus	292.36 °C	291.29 °C	−0.37
tout second condenser	165.65 °C	165.65 °C	0.00
tin second Claus	214.93 °C	214.93 °C	0.00
tout second Claus	222.58 °C	222.02 °C	−0.25
steam generated at the WHB	5257.24 kg/h	5249.81 kg/h	−0.14
H_2S at the tail gas	4106 ppmv	4162 ppmv	1.37
SO_2 at the tail gas	2336 ppmv	2360 ppmv	1.01
H_2S/SO_2 ratio	1.76	1.76	0.35
recovered sulfur		49.58976 t/day	
sulfur conversion		92.98547	

Table 7. Measured and Simulated Values for Test Run #4 Adopted in Implementing the EVM for Tuning the Adaptive Simulation

measures	test run #4: February 12, 2008, 9 am		
H_2O	0.0802 fraz mol		
CO_2	0.1015 fraz mol	AG	
H_2S	0.8184 fraz mol		
H_2O	0.2827 fraz mol		
H_2S	0.3027 fraz mol	SWS	
NH_3	0.4146 fraz mol		
total H_2S	49 t/day	49 t/day	
	measured	simulated	residuals
AG flow	2273.37 kg/h	2273.37 kg/h	0.00
AG temperature	54.41 °C	54.41 °C	0.00
SWS flow	396.38 kg/h	396.38 kg/h	0.00
SWS temperature	85.08 °C	85.08 °C	0.00
combustion air flow	5414.27 kg/h	4606.64 kg/h	−14.92
combustion air temperature	211.05 °C	211.05 °C	0.00
furnace temperature	1392.38 °C	1356.39 °C	−2.58
WHB temperature	309.07 °C	309.07 °C	0.00
tout first condenser	167.61 °C	167.61 °C	0.00
tin first Claus	229.30 °C	229.30 °C	0.00
tout first Claus	293.17 °C	289.83 °C	−1.14
tout second condenser	165.98 °C	165.98 °C	0.00
tin second Claus	215.21 °C	215.21 °C	0.00
tout second Claus	222.78 °C	220.11 °C	−1.20
steam generated at the WHB	5571.41 kg/h	4707.72 kg/h	−15.50
H_2S at the tail gas	4223 ppmv	3970 ppmv	−5.99
SO_2 at the tail gas	2371 ppmv	2263 ppmv	−4.56
H_2S/SO_2 ratio	1.78	1.75	−1.50
recovered sulfur		44.62848 t/day	
sulfur conversion		91.51679	

one gross error in the data set of test run #4. A deeper off-line analysis of data set showed that the mean square error[24,43] of test run #4 can be reasonably reduced by removing from the objective function the quadratic term dealing with the measured flow rate of sour water stripper (SWS), by pointing out that such a value is significantly wrong with respect to the other data. A posteriori, the presence of a gross error on the SWS line has been confirmed by field operators as shown in Figure 5. It is worth considering that the SWS line is a header receiving two flow rates, and the instrumentation failure has occurred on one of them only. The inferred measure of SWS line and the corresponding results are reported in Table 8.

Table 8. Measured and Simulated Values for Test Run #4 Adopted in Implementing the EVM for Tuning the Adaptive Simulation[a]

measures	test run #4: February 13, 2008, 9 am		
H_2O	0.0802 fraz mol		
CO_2	0.1015 fraz mol	AG	
H_2S	0.8184 fraz mol		
H_2O	0.2827 fraz mol		
H_2S	0.3027 fraz mol	SWS	
NH_3	0.4146 fraz mol		
total H_2S	52 t/day	52 t/day	
	measured	simulated	residuals
AG flow	2273.37 kg/h	2273.37 kg/h	0.00
AG temperature	54.41 °C	54.41 °C	0.00
SWS flow	735.51 kg/h	735.51 kg/h	0.00
SWS temperature	85.08 °C	85.08 °C	0.00
combustion air flow	5414.27 kg/h	5485.88 kg/h	1.32
combustion air temperature	211.05 °C	211.05 °C	0.00
furnace temperature	1392.38 °C	1389.03 °C	−0.24
WHB temperature	309.07 °C	309.07 °C	0.00
tout first condenser	167.61 °C	167.61 °C	0.00
tin first Claus	229.30 °C	229.30 °C	0.00
tout first Claus	293.17 °C	291.50 °C	−0.57
tout second condenser	165.98 °C	165.98 °C	0.00
tin second Claus	215.21 °C	215.21 °C	0.00
tout second Claus	222.78 °C	222.36 °C	−0.19
steam generated at the WHB	5571.41 kg/h	5178.90 kg/h	−7.05
H_2S at the tail gas	4223 ppmv	4184 ppmv	−0.92
SO_2 at the tail gas	2371 ppmv	2349 ppmv	−0.93
H_2S/SO_2 ratio	1.78	1.78	0.01
recovered sulfur		44.62848 t/day	
sulfur conversion		85.35633	

[a] A gross error was identified and corrected in correspondence with the SWS flow rate.

Facing the robustness of the proposed approach, it is worth highlighting that the adaptive simulation here proposed cannot automatically point out which specific measures are really wrong, but it can only show that the measures coming from the field are either coherent to each other or not by providing the closest (and numerically convergent) coherent picture of the plant according to the corresponding set of measures. It is still the task of the operator to investigate when deviations are significantly wrong. Obviously, before proceeding to an online application, there is the necessity to make automatic the detection and the correction of gross errors to make both simulation and reconciliation not affected by them.

Furthermore, taking into account the difference of the AG composition with respect to the previous four test runs, it is not a coincidence that the simulation of test run #5 (see Table 9) slightly underestimates the combustion air flow (about −5%) against the measured one; this induces a reduced steam generation in the waste heat boiler (about −700 kg/h and −11% of relative deviation), even though it is part of the EVM test run set. All the more so, the selected validation cases of Tables 10−12 show a few deviations for the combustion air flow (the simulated values are somewhat underestimated by few percents), hence, the steam generated at the waste heat boiler (about −10%). The deviations are due to the fact that the AG and SWS compositions are not measured, and the composition has been defined once on the process basis and used for all three simulation cases. Even though this simulation package is an effective tool able to give for all steady-state operating conditions a coherent picture of the plant during its own run length, this ultimate goal is to provide a reliable tool to reduce these deviations by inferring the composition with a coaptation procedure based on data reconciliation. Related issues shall be described in a future paper.

This ability of inferring data with a certain accuracy is essential to face the serious problem of missing data due to the lack of instrumentation; thanks to inferred data, performance

Table 9. Measured and Simulated Values for Test Run #5 Adopted in Implementing the EVM for Tuning the Adaptive Simulation

measures	test run #5: February 13, 2008, 1 pm		
H_2O	0.0841 fraz mol		
CO_2	0.1350	AG	
H_2S	0.7806 kmol/h		
H_2O	0.2908 fraz mol		
H_2S	0.2990	SWS	
NH_3	0.4100 kmol/h		
total H_2S	57 t/day	57 t/day	
	measured	simulated	residuals
AG flow	2724.62 kg/h	2724.62 kg/h	0.00
AG temperature	54.90 °C	54.90 °C	0.00
SWS flow	622.89 kg/h	622.89 kg/h	0.00
SWS temperature	85.17 °C	85.17 °C	0.00
combustion air flow	6005.18 kg/h	5732.47 kg/h	−4.54
combustion air temperature	207.09 °C	207.09 °C	0.00
furnace temperature	1386.06 °C	1357.23 °C	−2.08
WHB temperature	313.86 °C	313.86 °C	0.00
tout first condenser	168.10 °C	168.10 °C	0.00
tin first Claus	228.50 °C	228.50 °C	0.00
tout first Claus	294.41 °C	293.15 °C	−0.43
tout second condenser	166.04 °C	166.04 °C	0.00
tin second Claus	215.40 °C	215.40 °C	0.00
tout second Claus	222.63 °C	223.59 °C	0.43
steam generated at the WHB	6034.60 kg/h	5338.38 kg/h	−11.54
H_2S at the tail gas	3125 ppmv	3351 ppmv	7.23
SO_2 at the tail gas	3545 ppmv	3833 ppmv	8.14
H_2S/SO_2 ratio	0.88	0.87	−0.84
recovered sulfur		52.5312 t/day	
sulfur conversion		91.46677	

Table 10. Measured and Simulated Values for Test Run #6 Adopted in Validating the Simulation Adapted to the Industrial Case in Study

measures	test run #6: January 6, 2008, 6 am		
H_2S in AG flow	0.7913 fraz mol 71.759 kmol/h		
H_2S in SWS flow	0.300 fraz mol 14.381 kmol/h		
total H_2S	66 t/day	66 t/day	
	measured	simulated	residuals
AG flow	2901.81 kg/h	2901.81 kg/h	0.00
AG temperature	54.30 °C	54.30 °C	0.00
SWS flow	1073.77 kg/h	1073.77 kg/h	0.00
SWS temperature	85.20 °C	85.20 °C	0.00
combustion air flow	7342.26 kg/h	7136.50 kg/h	−2.80
combustion air temperature	220.58 °C	220.58 °C	0.00
furnace temperature	1387.93 °C	1388.74 °C	0.06
WHB temperature	313.76 °C	313.76 °C	0.00
tout first condenser	168.09 °C	168.09 °C	0.00
tin first Claus	227.17 °C	227.17 °C	0.00
tout first Claus	289.30 °C	294.03 °C	1.64
tout second condenser	164.92 °C	164.92 °C	0.00
tin second Claus	215.82 °C	215.82 °C	0.00
tout second Claus	221.87 °C	226.10 °C	1.91
steam generated at the WHB	7453.13 kg/h	6718.43 kg/h	−9.86
H_2S at the tail gas	4258 ppmv	4521 ppmv	6.18
SO_2 at the tail gas	2364 ppmv	2494 ppmv	5.52
H_2S/SO_2 ratio	1.80	1.81	0.63
recovered sulfur		60.33408 t/day	
sulfur conversion		91.2002	

Table 11. Measured and Simulated Values for Test Run #7 Adopted in Validating the Simulation Adapted to the Industrial Case in Study

measures	test run #7: January 7, 2008, 7 am		
H_2S in AG flow	0.7913 fraz mol 67.339 kmol/h		
H_2S in SWS flow	0.300 fraz mol 15.311 kmol/h		
total H_2S	63 t/day	63 t/day	
	measured	simulated	residuals
AG flow	2723.07 kg/h	2723.07 kg/h	0.00
AG temperature	54.97 °C	54.97 °C	0.00
SWS flow	1143.24 kg/h	1143.24 kg/h	0.00
SWS temperature	85.24 °C	85.24 °C	0.00
combustion air flow	7249.42 kg/h	6993.853 kg/h	−3.53
combustion air temperature	220.43 °C	220.43 °C	0.00
furnace temperature	1383.51 °C	1393.57 °C	0.73
WHB temperature	312.52 °C	312.52 °C	0.00
tout first condenser	168.39 °C	168.39 °C	0.00
tin first Claus	226.75 °C	226.75 °C	0.00
tout first Claus	287.89 °C	293.57 °C	1.97
tout second condenser	164.80 °C	164.80 °C	0.00
tin second Claus	216.10 °C	216.10 °C	0.00
tout second Claus	224.18 °C	226.02 °C	0.82
steam generated at the WHB	7292.18 kg/h	6601.39 kg/h	−9.47
H_2S at the tail gas	5022 ppmv	5232 ppmv	4.18
SO_2 at the tail gas	1759 ppmv	1863 ppmv	5.93
H_2S/SO_2 ratio	2.86	2.81	−1.65
recovered sulfur		58.4064 t/day	
sulfur conversion		92.0142	

Table 12. Measured and Simulated Values for Test Run #8 Adopted in Validating the Simulation Adapted to the Industrial Case in Study

measures	test run #8: January 8, 2008, 7 am		
H_2S in AG flow	0.7913 fraz mol 64.538 kmol/h		
H_2S in SWS flow	0.300 fraz mol 16.237 kmol/h		
total H_2S	62 t/day	62 t/day	
	measured	simulated	residuals
AG flow	2609.81 kg/h	2609.81 kg/h	0.00
AG temperature	55.50 °C	55.50 °C	0.00
SWS flow	1212.33 kg/h	1212.33 kg/h	0.00
SWS temperature	84.74 °C	84.74 °C	0.00
combustion air flow	7144.55 kg/h	6942.67 kg/h	−2.83
combustion air temperature	221.64 °C	221.64 °C	0.00
furnace temperature	1377.35 °C	1396.63 °C	1.40
WHB temperature	311.83 °C	311.83 °C	0.00
tout first condenser	167.37 °C	167.37 °C	0.00
tin first Claus	227.79 °C	227.79 °C	0.00
tout first Claus	287.51 °C	294.39 °C	2.39
tout second condenser	165.02 °C	165.02 °C	0.00
tin second Claus	215.22 °C	215.22 °C	0.00
tout second Claus	222.68 °C	225.01 °C	1.05
steam generated at the WHB	7087.49 kg/h	6564.25 kg/h	−7.38
H_2S at the tail gas	6102 ppmv	6348 ppmv	4.04
SO_2 at the tail gas	1161 ppmv	1207 ppmv	4.00
H_2S/SO_2 ratio	5.26	5.26	0.03
recovered sulfur		55.296 t/day	
sulfur conversion		89.13656	

monitoring of plant/process units is more reliable, operators can properly see what the current SRU conditions are, and the online data reconciliation problem becomes feasible even for SRUs.

7. Conclusions

This work represents the essential basis of an academic−industrial activity involving Politecnico di Milano and Chemprod Srl aimed at proposing a generalized approach to simulate SRUs and reconcile data acquired by the field. Many problems of different natures must be solved to achieve the final goal on SRUs, and some of them are properly solved by means of the proposed adaptive simulation and the EVM to tune parameters, by leaving the remaining open issues to future developments.

In details, this work proposed an adaptive simulation of SRU plant that includes basic models belonging to the libraries of a commercial process simulator (specifically PRO/II by SimSci-Esscor but any other process simulator could be used), some heuristic relations dictated by the industrial experience, and a set of adaptive parameters to make the detailed simulation framework a generalized approach. In fact, one could adapt the SRU simulation on different plants having similar layouts (the majority of existing Claus processes), but one could also continuously retune the simulation to match the evolving conditions of the same plant (e.g., monthly, to account for fouling factors).

The simulation tuning was carried out by adopting the so-called EVM, even though some other simplified methods bringing to reduced size of the resulting nonlinear programming were proposed and compared.

Finally, a period of 2 months of an industrial large-scale SRU operating in an Italian refinery has been selected to tune the adaptive simulation and to validate the proposed approach by showing encouraging results. The adapted simulation showed a high flexibility by providing coherent pictures of the plant apart from the operating conditions analyzed, but at the same time, it preserved its high level of detail by giving the opportunity to still exploit the robustness of a complex and detailed simulation to infer some missing measures in a reliable way. Concluding, the adaptive simulation could be used to tackle the lack of measurement and instrumentation that is typical of SRU plants and therefore to become the essential basis for a generalized robust and reliable data reconciliation tool for the online application, with which the next developments shall deal.

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