Modeling a Flue-Gas Desulfurization Plant with a Fuzzy Methodology to Optimize the SO$_2$ Absorption Process

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Abstract: Sulfur oxides are some of the major existing pollutants that directly affect the atmosphere. In combination with particles and air humidity, produce the most detrimental effects attributed to air pollution. The treatment of gas streams containing sulfur dioxide and its subsequent recovery is, therefore, a matter of great importance for the elimination of the environmental burden of their emission into the atmosphere. In this research, a fuzzy model of a flue-gas desulfurization plant is developed with the aim of dealing with two optimizations problems. The first one, is centered in finding the amount of liquid that should be injected into the plant in order to optimize the SO$_2$ absorption process. The second one, is the development of a tool to help to size the absorption tower (find the right dimension), given the optimum amount of liquid derived from the previous goal. The results obtained, although preliminary, are reliable and useful for chemical engineering plant design.

1 INTRODUCTION

The sources of emission of flue gas pollutants are varied, they come mainly from industrial processes of the petrochemical industry, energy industry as thermal plants, or in industries with its own system of generation of energy by means of boilers or gas turbines. Other sources of minority emissions come from the use of fossil fuels in transport or heating boilers in residential buildings.

Sulfur oxides are some of the major existing pollutants that directly affect the atmosphere. These include sulfur dioxide and sulfur trioxide.

Sulfur dioxide (SO$_2$) is a colorless gas with a strong, irritating and toxic odor that is usually contained in small traces of fossil fuels. The main source of SO$_2$ generation comes from the burning of fossil fuels and it is estimated that large industrial complexes account for approximately 90% of SO$_2$ emissions. Other sources of anthropological SO$_2$ may be metallurgical industrial processes, paper industry and the production of sulfuric acid.

Considering the physicochemical properties of SO$_2$ and the volume in which it is generated, residual SO$_2$ is considered as one of the most important threats to the environment.

Sulfur oxides in combination with particles and air humidity produce the most detrimental effects attributed to air pollution. It affects, among others, visibility (atmospheric haze), materials (metal corrosion, tissue deterioration, etc.) and health (respiratory system irritation and chronic respiratory problems), vegetation (necrosis in plants). It also produces the acid rain, which can be caused by dry deposition (gas and particle settling from the atmosphere) and wet deposition (acid rain, fog and snow), causing a phenomenon of acidification of natural sources of water and leaching of soil nutrients.

It is also important to note that sulfur oxides, together with other pollutants such as nitrogen oxides and particulates, cause transboundary air pollution, and their emission into the atmosphere is a matter of global concern.

The treatment of gas streams containing sulfur dioxide and its subsequent recovery is, therefore, a matter of great importance for the sustainable management of resources and the elimination of the environmental burden of their emission into the
At present Flue-Gas Desulfurization (FGD) is commonly used in industrial processes to control SO2 emissions. Within these processes we can find numerous techniques including wet (aqueous solution) and dry (limestones) scrubbers, adsorption systems (active carbon) and catalytic or regeneration systems.

Catalytic systems allow the valorisation of SO2, obtaining by-products (such as sulfuric acid or elemental sulphur) which can be exploited as a raw material. These technologies are based on a first stage where SO2 is transferred from air current to an aqueous solution.

In these research, a wet scrubber included in an elemental sulphur recovery process is presented, from which data have been generated to model its behavior.

The modeling is performed using the methodology of the Fuzzy Inductive Reasoning (FIR) with a double objective. On the one hand, study the amount of liquid that must be injected into the plant in order to optimize the SO2 absorption process. On the other hand, it is tried to size the absorption tower, looking for the best Empty Bed Residence Time (EBRT) given the amount of optimum liquid found in the previous objective.

Section 2 presents the flue-gas desulfurization plant designed and developed. The FIR methodology chosen for this study is introduced in Section 3. Section 4 presents the experimentation and the results obtained for the two objectives proposed. Finally, a discussion and the conclusions are presented.

2 THE FLUE-GAS DESULFURIZATION (FGD) PLANT

The FGD plant is based on wet absorption carried out on a spray column. The scrubber consisted of a 6 cm diameter and 30 cm height column, and an 11 cm diameter and 20 cm height liquid reservoir.

Fresh liquid phase was continuously fed (without recirculation) assuring a constant composition. Conductivity and pH were monitored in the liquid effluent. The concentration of SO2 in the air streams was monitored using a selective electrochemical sensor. The FGD plant designed and developed is presented in Figure 1.

The FGD plant was operated as a spray column since this type of scrubber presents several advantages. This type of column is the simplest and most robust because it does not contain a packing material to favours mass transfer. Therefore, this system is the low loss of pressure produced with respect to other available absorption systems.

The transfer of SO2 from gas phase to liquid phase is the key factor in this process, limiting the removal efficiency (RE). For this reason, mass transfer between both phases is favoured in spray columns by increasing the interface area and thereby the mass transfer rate (Green and Perry, 2008). With this purpose liquid is dispersed into the polluted gas as small droplets (as shown in Figure 2).

In the spray column presented herein, liquid phase was continuously injected into the column, using a spray-nozzle, as 50 μm droplets. At the same time, the SO2 rich-gases were fed counter-currently to improve the SO2 transfer mechanisms (Sinnot, 2012).

Considering SO2 solubility in aqueous solutions, water was selected as absorbent solution due to its high availability and low cost (Kohl and Nielse, 1997). The absorption of SO2 into water occurs following these equations:

\[
SO_2(g) + H_2O(l) \leftrightarrow HSO_3^-(aq) + H^+(aq)
\]

\[
HSO_3^-(aq) \leftrightarrow SO_3^{2-}(aq) + H^+(aq)
\]

By these reactions, SO2 was simply removed from the polluted air stream, forming an acidic solution residue of the absorption.

- Figure 1: FGD plant picture.
3 THE FUZZY INDUCTIVE REASONING (FIR) METHODOLOGY

The conceptualization of the Fuzzy Inductive Reasoning (FIR) methodology arises from the General Systems Theory (GSPS) proposed by Klir (1969). This modeling and qualitative simulation methodology is based on systems behavior rather than on structural knowledge. It is able to obtain good qualitative relations between the variables that compose the system and to infer the future behavior of that system. It also has the ability to describe systems that cannot easily be described by classical mathematics (e.g. differential equations), i.e. systems for which the underlying physical laws are not well understood. FIR structure is schematically presented in Figure 3.

FIR consists of four main processes, namely: fuzzification, qualitative model identification, fuzzy forecast and defuzzification. The fuzzification process converts quantitative data stemming from the system into fuzzy data, which consist of a triplet containing the class, the membership and side values (Nebot et al., 2012; 2010). The qualitative model identification process is responsible for finding causal and temporal relations between variables and therefore for obtaining the best model that represents the system.

A FIR model is composed of a mask (model structure) and a pattern rule base (behaviour matrix). An example of both is presented in Figure 4.

The qualitative model identification process evaluates which mask has the highest prediction power by means of an entropy reduction measure, called the quality \( Q \) of the mask. The mask with the maximum \( Q \) value is the optimal one. Once the best mask has been identified, it can be applied to the qualitative data obtained from the system resulting in a particular fuzzy pattern rule base, also called behavior matrix in FIR nomenclature (see Figure 4).

Once the FIR model is available, the prediction can take place using the FIR inference engine, called fuzzy forecast process, which is a specialization of the \( k \)-nearest neighbor rule commonly used in the pattern recognition field. In this research a \( k = 5 \) is chosen. Finally, defuzzification is the inverse process of fuzzification. It allows converting the qualitative predicted output into quantitative values that can then be used as inputs to an external quantitative model. For a deeper insight into FIR methodology the reader is referred to (Nebot et al., 2012; 2010).

3.1 The Concept of Mask in FIR: Feature Selection Process

In FIR, a mask candidate matrix is the ensemble of all possible masks. The mask candidate matrix contains elements of value -1, where the mask has a potential m-input (mask input), a value +1 where the mask has its m-output (mask output), and a 0 value to denote forbidden connections. Each negative element in the mask denotes a possible causal relation with the output. A good mask candidate matrix to determine a
predictive model for a system with four input and one output variables is shown in Figure 5.

<table>
<thead>
<tr>
<th>x</th>
<th>t</th>
<th>u1</th>
<th>u2</th>
<th>u3</th>
<th>u4</th>
<th>y1</th>
</tr>
</thead>
<tbody>
<tr>
<td>t-2δt</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td></td>
</tr>
<tr>
<td>t-δt</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td></td>
</tr>
<tr>
<td>t</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>+1</td>
<td></td>
</tr>
</tbody>
</table>

Figure 5: Example of mask candidate matrix. t-δt means a time stamp in the past and t-2δt two time stamps in the past.

Starting from the candidate matrix the qualitative model identification process searches through all legal masks of complexity two, i.e. all masks with a single m-input and finds the best one; it then proceeds by searching through all legal masks of complexity three, i.e. all masks with two m-inputs and finds the best of those; and it continues in the same manner until the maximum complexity has been reached. This strategy corresponds to an exhaustive search of exponential complexity.

Each of the possible masks is compared to the others with respect to its forecasting power, which is maximal when the associate entropy measure is minimal. The Shannon entropy measure is used to determine the uncertainty associated with forecasting a particular output state given any legal input state. The Shannon entropy relative to one input state is calculated using the Equation presented in (1).

\[ H_i = \sum_{o} p(o|i) \cdot \log_2 p(o|i) \]  \hspace{1cm} (1)

where \( p(o|i) \) is the conditional probability of a certain m-output state \( o \) to occur, given that the m-input state \( i \) has already occurred. It denotes the quotient of the observed frequency of a particular state divided by the highest possible frequency of that state. The overall entropy of the mask is then computed as the weighted sum of the entropy over all input states (Equation 2).

\[ H_m = -\sum_{vi} p(i) \cdot H_i \]  \hspace{1cm} (2)

where \( p(i) \) is the probability of that input state to occur. The highest possible entropy \( H_{max} \) is obtained when all probabilities are equal, and zero entropy corresponds to totally deterministic relationships. A normalized overall entropy reduction \( H_r \) is defined as described in Equation 3.

\[ H_r = 1.0 \left( 1 - \frac{H_m}{H_{max}} \right) \]  \hspace{1cm} (3)

\( H_r \) is a real-valued number in the range between 0.0 and 1.0, where high values indicate an improved forecasting power.

From a statistical point of view, every state should be observed at least five times (Law and Kelton, 1991). Therefore, an observation ratio, \( O_r \), is introduced as an additional contributor to the overall quality measure:

\[ O_r = \frac{5 \cdot n_{5x} + 4 \cdot n_{4x} + 3 \cdot n_{3x} + 2 \cdot n_{2x} + n_{1x}}{5 \cdot n_{log}} \]  \hspace{1cm} (4)

where: \( n_{log} \) is the number of legal m-input states, \( n_{1x} \) is the number of m-input states observed only once, \( n_{2x} \) is the number of m-inputs states observed twice, and so on. The overall quality of a mask, \( Q \), is then defined as the product of its uncertainty reduction measure, \( H_r \), and its observation ratio, \( O_r \):

\[ Q = H_r \cdot O_r \]  \hspace{1cm} (5)

An example of a mask is presented in Figure 6.

As mentioned before, each negative element in the mask is called an m-input (mask input). It denotes a causal and temporal relation with the output, i.e. it influences the output up to a certain degree. The enumeration of the m-inputs is immaterial and has no relevance. Let us now address the second issue related to the model identification process of FIR methodology. How is the pattern rule base obtained from the mask? This process is illustrated in Figure 4. The mask can be used to ‘flatten’ dynamic relationships into pseudo-static relationships. The left side of Figure 4 shows an excerpt of the qualitative data matrix that stores the class values. The dashed box symbolizes the mask that is shifted downwards along the class value matrix. The round shaded ‘holes’ in the mask denote the positions of the m-inputs, whereas the square shaded ‘hole’ indicates the position of the m-output. The class values are read out from the class value matrix through the ‘holes’ of the mask, and are placed next to each other in the behaviour matrix that is shown on the centre of Figure 4. Here, each row represents one position of the mask along the class value matrix. It is lined up with the bottom row of the mask. Each row of the behaviour matrix represents one pseudo-static qualitative state.
or qualitative rule (also called fuzzy pattern rule) (Nebot et al., 2012; 2010).

4 EXPERIMENTS AND RESULTS

As mentioned previously, in this research it was studied the use of the FIR methodology to model the FGD plant. From the model obtained, two important aspects of the proper plant were optimized. The main goals addressed in this work are twofold:

- To find the amount of liquid that should be injected into the plant in order to optimize the SO2 absorption process.
- To develop a tool to help to find the right dimension of the absorption tower, by obtaining the best EBRT given the optimum amount of liquid derived from the previous goal.

This section addresses the modelling of both goals and presents the results obtained.

4.1 Experimental Data Set

The set of data available for this study has been obtained from the FGD plant presented in section 2. The first 9 parameters described in Table I correspond to the input parameters of the plant, whereas the output parameter is in the last row of the table, i.e. the amount of SO2 obtained after the desulfurization process.

A set of 269 experiments have been performed changing the values of some of the input parameters described in Table I, such are QL, QG, SO2I and pH.

<table>
<thead>
<tr>
<th>QL</th>
<th>Liquid injected into the plant</th>
</tr>
</thead>
<tbody>
<tr>
<td>QG</td>
<td>Gas injected into the plant</td>
</tr>
<tr>
<td>pH</td>
<td>Initial pH</td>
</tr>
<tr>
<td>SO2I</td>
<td>Initial SO2 (SO2 at the beginning of the cleaning process)</td>
</tr>
<tr>
<td>SO42</td>
<td>Sulfate</td>
</tr>
<tr>
<td>TLIQUID</td>
<td>Liquid temperature</td>
</tr>
<tr>
<td>TGAS</td>
<td>Gas temperature</td>
</tr>
<tr>
<td>EBRT</td>
<td>Empty Bed Residence Time (meaning volume of biofilter divided by the incoming airflow)</td>
</tr>
<tr>
<td>L/G</td>
<td>Relation between the input liquid and the gas</td>
</tr>
<tr>
<td>SO2F</td>
<td>Final SO2 (SO2 at the end of the desulfurization process)</td>
</tr>
</tbody>
</table>

Table I: Parameters of the plant.

With this three input variables it is possible to predict quite accurately the main behaviour of the plant.

This FIR model has been validated by predicting a subset of the available data, not used in the training process, getting a Mean Square Error (MSE) measure in percentage of 10.4%, that is an acceptable result.

Once a validated FIR model is available, the next step is to try to optimize two important aspects of the FGD plant: 1) the amount of liquid that should be injected into the plant in order to optimize the SO2 absorption process; 2) the right dimension (size) of the absorption tower, by obtaining the best EBRT given the optimum amount of liquid derived from the previous goal.

4.2 FIR Model of the FGD Plant

The next step is to find a FIR model that describes as much accurately as possible the behavior of the plant extracting the knowledge from the data set available.

Notice, that each data instance represents a specific plant condition measurement and its corresponding final SO2 obtained when the desulfurization process is finished. Therefore, each instance is time independent from each other. That means that the FIR mask candidate matrix has, in this research, only one row, since no temporal relations should be studied between the variables involved.

All variables in this research (described in Table I), have been discretized into three classes, using different discretization algorithms, i.e. EFP (equal frequency partition), EWI (equal with interval), etc., depending on the specific characteristics of the data.

It should be mentioned here, that variables SO42 and TGAS contain missing values in almost all their measurements, therefore, they have been removed from the modeling.

The qualitative identification process of the FIR methodology has been used to obtain the mask (model structure) and the pattern rule base (set of rules containing system’s behavior). The mask obtained with the highest quality is presented in Figure 7.

As can be seen from the mask shown in Figure 7, FIR encountered that the most relevant variables involved in the prediction of the final SO2 are the liquid injected into the plant (QL), the initial SO2 (SO2I) and the empty bed residence time (EBRT).

With this three input variables it is possible to predict quite accurately the main behaviour of the plant.

This FIR model has been validated by predicting a subset of the available data, not used in the training process, getting a Mean Square Error (MSE) measure in percentage of 10.4%, that is an acceptable result.

Once a validated FIR model is available, the next step is to try to optimize two important aspects of the FGD plant: 1) the amount of liquid that should be injected into the plant in order to optimize the SO2 absorption process; 2) the right dimension (size) of the absorption tower, by obtaining the best EBRT given the optimum amount of liquid derived from the previous goal.
4.3 **Amount of Liquid Injected into the Plant**

The objective in this experiment is to look for the amount of liquid that must be injected into the FGD plant in order to optimize the process of absorption of the sulphur dioxide (SO₂).

With this goal in mind, the FIR model of the FGD plant, described in the previous section, is used as shown in Figure 8, to predict SO₂F. In this experiment, the EBRT value is set to 0.8 seconds and for different initial values of SO₂ (SO₂I), the optimum amount of liquid is found, i.e. the amount of liquid to be injected into the plant, which gives less sulphur dioxide at the exit.

As can be seen in Figure 8, the recode process of FIR methodology is applied to each of the three relevant variables encountered by the mask, i.e. QL, SO₂I and EBRT. The input variables, already converted from quantitative to qualitative values, are the inputs of the FIR prediction process, which uses the mask of Figure 7 and the pattern rule base extracted out of it, to perform the prediction. Finally, the predicted SO₂F value is converted again to a quantitative value by means of the regeneration process of FIR.

The simulation results obtained for this experiment, given an SO₂I value of 4000 ppmv, are shown in Figure 9. The upper plot of Figure 9 presents the final SO₂, given an initial SO₂ of 4000 ppmv. The lower plot shows the increment applied to the QL variable during simulation, that ranges from 14 l/h to 26 l/h.

As can be seen from this simulation, the lowest SO₂F value is obtained when the amount of liquid injected into the plant is of almost 19 l/h.

4.4 **Right Dimension of the Absorption Tower**

The goal of this experiment is to develop a tool to help to find the right dimension (size) of the absorption tower, by obtaining the best empty bed residence time (EBRT) given the optimum amount of liquid derived from the previous goal, i.e. 18 l/h.

![Figure 8: Simulation experiment of the amount of liquid injected into the plant using SIMULINK. EBRT, QL and SO₂I input variables are discretized by means of the recode function and the FIR prediction module is used to obtain the predicted values that are then regenerated by using the regeneration function of FIR.](image)

Figure 8 shows the mesh obtained when several simulations are performed using different initial values of SO₂.

![Figure 9: SO₂F prediction results obtained given an SO₂I value of 4000 ppmv, an EBRT of 0.8 seconds and different QL values.](image)

In Figure 10, it can be seen that for SO₂I values less than 5000 ppmv, a minimum QL value of around 18 l/h is obtained in each curve. For higher SO₂I values, the model found predicts that the amount of SO₂ that is absorbed does not depend on the liquid injected into the plant.
With this new goal in mind, the FIR model of the FGD plant, described in the previous section, is used as shown in Figure 11, to predict SO$_2$F.

In this experiment, the QL value is set to 18 l/h (optimal value obtained in the previous experiment), and for different initial values of SO$_2$ (SO$_2$I), the optimum value of EBRT is found, i.e. the dimension of the absorption tower, which gives less sulphur dioxide at the exit. The same comments of Figure 8 apply to Figure 11.

Two examples of the simulation results obtained for this experiment are shown in Figures 12 and 13.

In Figure 12 the SO$_2$F prediction results obtained by the FIR model for an initial SO$_2$ of 2000 ppmv, a QL of 18 l/h, and changing the EBRT values from 0 to 100 seconds, are presented.

Figure 13 shows the SO$_2$F prediction results in the same conditions except that now the SO$_2$I is set to 4000 ppmv.

For an initial SO$_2$I value of 2000 ppmv, the optimal EBRT value is 2.48 seconds, whereas for a value of 4000 ppmv, the optimal EBRT is increased until a value of 3.84 second. Notice that EBRT represents the time that the gas is inside the column.

Figure 14 shows the mesh obtained when several simulations are performed using different values of SO$_2$I.
obtained starting from a set of 269 experiments performed in a real FGD plant that uses a spray column. The set of registered experiments have been performed changing the values of some of the input parameters, such as QL, QG, SO2I and pH.

It is quite clear that 269 experiments is not enough data to obtain a whole accurate and reliable FGD plant model. However, in this work it has been demonstrated that FIR model is able to capture the knowledge of part of the space defined by the input parameters in an efficient way and use it to optimize several aspects of the FGD plant.

In this work two optimizations goals have been achieved. The first one, is centered in finding the amount of liquid that should be injected into the plant in order to optimize the SO2 absorption process. The second one, is the development of a tool to help to find the right dimension of the absorption tower, by obtaining the best EBRT given the optimum amount of liquid derived from the previous goal.

The results obtained are useful for chemical engineering plant design and future developments involving more data experiments will be performed.

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