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# Modelling systems defined by RTD curves

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## ABSTRACT

The RTD theory is commonly used for describing flow patterns in a large class of applications, and particularly for ventilated enclosures. Experimental RTD curves are used for modelling these premises with an application in the nuclear industry for predicting the airborne pollutant transfers in order to prevent radiological risk. An approach based on a superstructure involving interlinked elementary flow patterns such as CSTRs, PFRs, recycles and by-passes is implemented. In order to propose a generic and easy to use tool, the associated large-scale MINLP problem is solved by using the GAMS package. After a validation phase on examples with known solutions, a laboratory enclosure, called MELANIE, used in the nuclear industry is modelled. The comparison between experimental RTD curve and the ones obtained from models extracted from superstructures shows good agreement. The superstructure-based solution procedure constitutes an efficient and intermediate way between numerical simulations using CFD codes and experimental determinations of characteristic parameters, which are both difficult to implement in the case of large and cluttered systems which are typical of the nuclear industry.

## 1. Introduction

In many industrial premises, particularly in the nuclear industry to prevent the radiological risk, the control of the airborne contamination transfers is an essential feature for operator protection and facility safety. Since the ventilation constitutes a widely used system of contamination containment, tools to assess airborne contamination transfers inside a ventilated enclosure have been developed by many researchers. These works are mainly based on the one hand on numerical simulation using computational fluid dynamics (CFD), and on the other hand on experimental determination of characteristic parameters. In real world applications, the experimental approach is generally difficult to implement, because a great number of interdependent parameters have to be estimated, so the results are often not representative of the physical phenomenon. In the case of large and cluttered systems which are the scope of this study, the predictive methods based on CFD codes would require a too thin meshing of the premise for obtaining results with an acceptable level of accuracy.

*Abbreviations:* CAD, computer aided design; CSTR, continuous stirred tank reactor; GAMS, general algebraic modelling system; PFR, plug flow reactor; CFD, computational fluid dynamics; MINLP, mixed-integer nonlinear programming; RTD, residence time distribution.

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This paper proposes an intermediate way between the purely numerical approach and the experimental characterization of the contamination transfers. The method lies on the determination of the experimental residence time distribution (RTD) curve, generally obtained through the response of the system to a tracer release. RTD theory (Danckwerts, 1953) is commonly used to describe the flow patterns in a wide variety of applications. Then a model is built up from a combination of elementary ideal flow patterns such as perfect mixing (CSTR), plug flow (PFR), by-pass and recycling (Levenspiel, 1972). Each elementary cell is characterized by specific parameters such as mean residence time and volume. The adjustment of the model is derived from the comparison between the simulated response of the model to a stimulus and the experimental RTD curve. The parameters of various candidate structures are optimized, in terms of volumes and flow rates, in order to fit as well as possible the experimental curve.

In most of the previous works, only softwares performing the identification of a little number of parameters for models of which structures are provided by the user were available (Berne & Blet, 1998; Leclerc, Detrez, Bernard, & Schweich, 1995; Thereska, 1998). Until the work of Laquerbe, Laborde, Soares, Floquet, et al. (2001), no software allowing a complete computer aided design of this type of structures has already been developed. The establishment of a RTD model lies first in the experimental determination of the residence time distribution curve, obtained generally through the response of the system to a tracer release, classically helium in ventilation applications. Then the simulated response of a proposed model to

## Nomenclature

$C_{exp}^k$	experimental concentration at step time $k$
$C_{exp}(t)$	experimental concentration at time $t$
$C_i^k$	concentration of elementary cell $i$ at step time $k$
$C_{in}^k$	input concentration at step time $k$
$C_{mod}^k$	concentration obtained from the model at step time $k$
$C_{neg}^k$	positive real variable at step time $k$
$C_{out}^k$	output concentration at step time $k$
$C_{pos}^k$	positive real variable at step time $k$
DZ	dead zone
$F_{CSTR}$	input flow rate of a CSTR
$F_i$	flow rate associated with elementary cell $i$
$F_{in}$	input flow rate on the network
$F_{PFR}$	input flow rate of a PFR
$h$	step time
$k_{max}$	number of discretization points in the time space
$M$	positive constant
$s$	sigmoid function
$t^k$	time at step $k$
$t$	time
$T_{neg}^k$	positive real variable at step time $k$
$T_{pos}^k$	positive real variable at step time $k$
$V_{CSTR}$	volume of a CSTR
$V_i$	volume of reactor $i$
$V_{PFR}$	volume of a PFR
$V_{Total}$	total volume of the enclosure
$y_i$	binary variable associated with elementary cell $i$
$z^k$	binary variable at step time $h$

### Greek letters

$\alpha$	positive constant
$\rho$	penalty coefficient

### Indexes

in	input
$k$	step time
out	output
mod	model
exp	experimental

a stimulus is compared with the experimental curve in order to test if the model fits well the experimental conditions.

As suggested by [Walter and Pronzato \(1994\)](#), the definition of a RTD model can be characterized as follows:

- modelling is performed in differed time;
- structures of the models are variable;
- the model is essentially a compartmental one, but constraints can be introduced to provide a phenomenological significance.

The problem involves two interlinked sub-problems: the structural identification of the models and the parametric identification of each proposed model. So the problem makes part of the MINLP optimization problem class, insofar as discrete variables related to the model structures (number and type of model components) and continuous ones translating operating conditions (volumes, flow rates) of the various model items simultaneously appear in the problem formulation.

In the chemical engineering field two classes of methods for solving MINLP problems can be distinguished. The determinis-

tic way, where the MINLP problem is solved by means of the generalized Bender's decomposition ([Yuan, Zhang, Pibouleau, & Domenech, 1988](#)), or the outer-approximation/equality-relaxation (OA/ER) procedure, first proposed by [Duran and Grossmann \(1986\)](#), [Kocis and Grossmann \(1987\)](#), and improved by [Kocis and Grossmann \(1988\)](#), or the branch and bound procedure ([Gupta, 1980](#); [Gupta & Ravindram, 1981](#)). In the other class of methods, the stochastic ones, a two-level optimization method is carried out ([Laquerbe, Laborde, Soares, Floquet, et al., 2001](#); [Laquerbe, Laborde, Soares, Ricciardi, et al., 2001](#); [Montastruc, Azzaro-Pantel, Pibouleau, & Domenech, 2004](#)). At the upper level the model structure is determined from a stochastic procedure, like the simulated annealing method or a genetic algorithm, and the operating conditions of the model units are optimized with a classical NLP code at the lower level.

In the previous study on ventilated enclosures of [Laquerbe, Laborde, Soares, Ricciardi, et al. \(2001\)](#), the model was built step by step by merging elementary patterns series cascade, recycling loop, parallel distribution, intermediate distribution of ideal components: CSTR, PFR, Short-circuit, and the resulting MINLP problem was solved by using a stochastic method. An alternative method, where on the one hand the model is searched within a pre-defined superstructure, and on the other hand the MINLP problem is solved by means of a deterministic procedure is proposed in this study.

The paper is organized as follows. In the first section, the problem of searching for an optimal solution within a superstructure composed of CSTR, PFR, recycles and by-passes, is formulated. The balance equations and the relations translating the existence or not an elementary unit give birth to a mixed-integer nonlinear programming problem (MINLP). Based on a previous study of [Hocine \(2006\)](#), the following part is related to the choice of a GAMS solver, namely the package SBB. In the fourth section, the approach combining the superstructure-based approach with the solver SBB is validated on two examples of which solution are known. In the fifth part, a real ventilated enclosure, the laboratory enclosure MELANIE is modelled for a step-up stimulus carried out by a helium release. Several numerical studies are performed and the results are compared with the one obtained by [Laquerbe, Laborde, Soares, Ricciardi, et al. \(2001\)](#), where the solution was built step by step by assembling elementary patterns. Finally, the significant contributions of this study and its potential use for modelling the dynamic behaviour of other chemical devices are summarized in a concluding part.

## 2. Problem formulation

For the synthesis of reactor networks, two solution approaches can be distinguished. The solving strategy in which the network is built step by step without embedding the set of potential solutions within a superstructure, was proposed by [Athier, Floquet, Pibouleau, and Domenech \(1997\)](#) and [Laquerbe, Laborde, Soares, Floquet, et al. \(2001\)](#). In these two-step procedures, the master problem, solved by a stochastic method like simulated annealing or a genetic algorithm, proposes network structures to the slave problem, where the continuous operating variables on the network under consideration (generally flow rates, concentrations and volumes) are optimized by a NLP method, QP or SQP according to the type of constraints. However, in this strategy the infeasibility of some structures proposed by the stochastic procedure has to be detected. This detection, lying on physical concepts is strongly linked to the problem under consideration, and suffers of a lack of applicability. Furthermore, the solutions provided by a stochastic procedure are without any doubt good solutions, but they are not optimal ones. That are the reasons why a procedure based on a

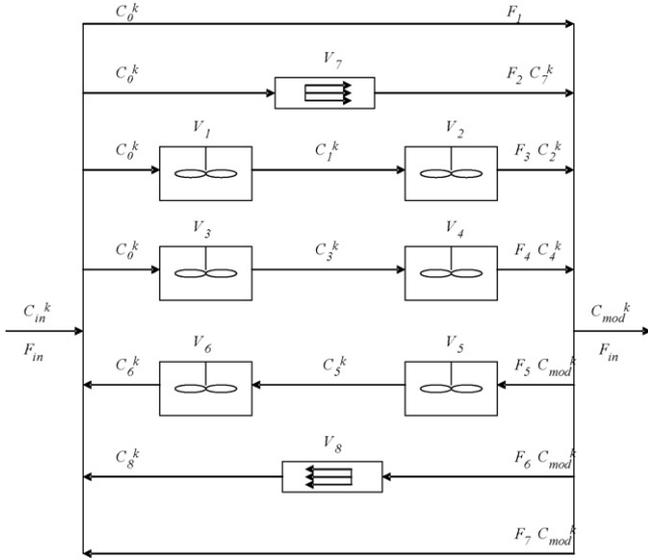


Fig. 1. Superstructure of the problem.

superstructure approach and a deterministic MINLP algorithm have been retained in this study.

The superstructure proposed by Hocine (2006) is used for searching the best model according to an experimental RTD curve. It involves six CSTR, two PFR, a by-pass and a recycle stream (see Fig. 1), that is to say 10 elementary modules, leading to a MINLP problem involving  $2^{10} = 1024$  potential solutions. In the case where the flow rates passing through an elementary cell are null, the cell is considered as a dead-zone. The binary variable  $y_i$  associated with each elementary pattern translate the absence or not of the pattern. The continuous variables for each elementary module  $i$  are the flow rate  $F_i$ , the concentration  $C_i^k$  where the index  $k$  refers to discretization step in the time interval (see below) and for a reactor, its volume  $V_i$  (Figs. 2a and 3b).

The mathematical formulation of the problem involves the following equations.

- Volume constraint:

$$\sum_{i=1}^8 V_i = V_{\text{Total}} \quad (1)$$

- Input node constraint:

$$F_{\text{in}} + F_1 + F_2 + F_3 + F_4 = F_5 + F_6 + F_7 \quad (2a)$$

$$C_{\text{in}}^k F_{\text{in}} + C_6^k F_5 + C_8^k F_6 + C_{\text{mod}}^k F_7 = C_0^k \sum_{i=1}^4 F_i \quad (2b)$$

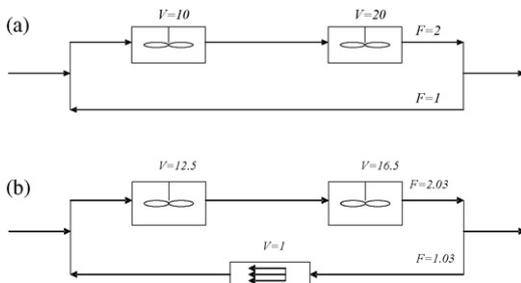


Fig. 2. (a) Reference model; (b) solution for a step-up stimulus.

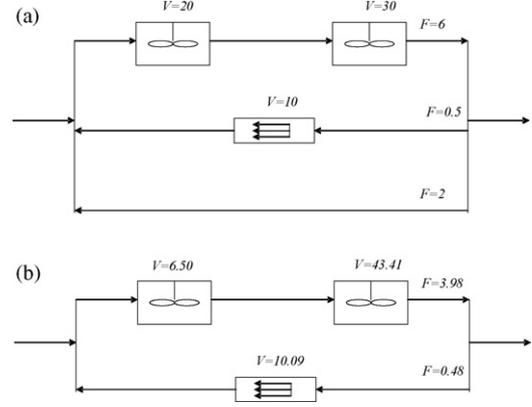


Fig. 3. (a) Reference model; (b) solution for a step-up stimulus.

- Output node constraint:

$$C_{\text{mod}}^k \sum_{i=5}^7 F_k = C_0^k F_1 + C_7^k F_2 + C_2^k F_3 + C_4^k F_4 \quad (3)$$

- Existence of reactors:

$$0 \leq V_i \leq y_i V_{\text{Total}} \quad (4)$$

- Existence of flows (for example flow 3):

$$F_3 \leq M(y_1 + y_2) F_{\text{in}} \quad (5)$$

This constraint expresses the flow rate existence, but also bounds the value of this flow rate. After numerous numerical trials, the value 10 was assigned to  $M$ .

- Bounds on variables:

$$0 \leq C_i^k \leq 1, \quad i = 0, 8 \quad (6a)$$

$$0 \leq C_{\text{mod}}^k \leq 1 \quad (6b)$$

$$0 \leq V_i \leq V_{\text{Total}} \quad (6c)$$

$$0 \leq F_i \leq M F_{\text{in}}, \quad i = 1, 4 \quad (6d)$$

$$0 \leq F_i \leq \frac{M}{2} F_{\text{in}}, \quad i = 5, 6, 7 \quad (6e)$$

Eqs. (6d) and (6e) are used to bound the search space for the flow rates. As in the previous case, the value  $M = 10$  was chosen. So in the direct sense it is assumed that each flow rate cannot be larger than 10 times as the input flow rate, and for the reverse sense this value is reduced to five.

In addition to this general set of constraints, specific equations have to be added for reactors.

- PFR reactors:

For a step-up stimulus, the PFR output concentration  $C_{\text{out}}$  is expressed in terms of its input concentration  $C_{\text{in}}$  by

$$C_{\text{out}} = 0 \quad \text{if} \quad t < \frac{V_{\text{PFR}}}{F_{\text{in,PFR}}} \quad (7a)$$

$$C_{\text{out}} = C_{\text{in}} \quad \text{if} \quad t \geq \frac{V_{\text{PFR}}}{F_{\text{in,PFR}}} \quad (7b)$$

At each step time  $k$ , these equations are approximated by the following continuous function derived from the classical sigmoid function (noted  $s$ ):

$$C_{\text{out}}^k = C_{\text{in}}^k \times s \left[ \alpha \left[ t^k - \frac{V_{\text{PFR}}}{F_{\text{in,PFR}}} \right] \right] \quad (7c)$$

where  $\alpha$  is a coefficient used to adjust the slope of the curve. The value 100 has been assigned to  $\alpha$ .

For an impulse stimulus, the PFR output is given by

$$C_{\text{out}} = 0 \quad \text{if } t \neq \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \quad (8a)$$

$$C_{\text{out}} = C_{\text{in}} \quad \text{if } t = \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \quad (8b)$$

In that case, the representation of the PFR output by means of a continuous mathematical function is rather difficult. To overcome this difficulty, the PFR response is defined according to a set of binary and continuous variables at each step time  $k$ . Let us define:

$$t^k - \frac{V_{\text{PFR}}}{F_{\text{PFR}}} = T_{\text{pos}}^k - T_{\text{neg}}^k \quad (8c)$$

with the constraints

$$C_{\text{out}}^k \leq 1 - z^k \quad (8d)$$

$$C_{\text{pos}}^k - C_{\text{neg}}^k \leq 2z^k \quad (8e)$$

$$T_{\text{pos}}^k + T_{\text{neg}}^k \leq \left[ \text{Max} \left( t^k - \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \right) \right] z^k \quad (8f)$$

$$C_{\text{out}}^k - C_{\text{in}}^k = C_{\text{pos}}^k - C_{\text{neg}}^k \quad (8g)$$

where  $z^k$  is a binary variable, and  $C_{\text{pos}}^k, C_{\text{neg}}^k, T_{\text{pos}}^k, T_{\text{neg}}^k \in \mathbb{R}^+$ .

If  $z^k = 0$ , it comes:

$$C_{\text{pos}}^k = C_{\text{neg}}^k \quad (8h)$$

$$C_{\text{out}}^k = C_{\text{in}}^k \quad (8i)$$

$$T_{\text{pos}}^k = T_{\text{neg}}^k = 0 \quad (8j)$$

$$t^k = \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \quad (8k)$$

In the same way, if  $z^k = 1$ , we have:

$$C_{\text{out}}^k = 0 \quad (8l)$$

$$T_{\text{pos}}^k + T_{\text{neg}}^k \leq \text{Max} \left( t^k - \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \right) \quad (8m)$$

$$t^k - \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \leq \text{Max} \left( t^k - \frac{V_{\text{PFR}}}{F_{\text{PFR}}} \right) \quad (8n)$$

#### • CSTR reactors:

The output of a CSTR is expressed as a function of its input by the classical ODE equation:

$$\frac{dC_{\text{out}}}{dt} = \frac{F_{\text{CSTR}}}{V_{\text{CSTR}}} \times (C_{\text{in}} - C_{\text{out}}) \quad (9a)$$

By using the classical Euler's method, it comes:

$$C_{\text{out}}^{k+1} = \left( \frac{hF_{\text{CSTR}}}{V_{\text{CSTR}}} \right) \times (C_{\text{in}}^k - C_{\text{out}}^k) + C_{\text{out}}^k \quad (9b)$$

where  $h$  is the step time,  $h = t^{k+1} - t^k$ .

According to the set of constraints above described, the MINLP problem consists in extracting from the superstructure shown in Fig. 1, the model fitting as well as possible an experimental RTD curve, discretized in  $k_{\text{max}}$  equidistant points in the time space. In addition to the variables defined at each step time ( $C_i^k, i = 0$  to  $8, C_{\text{mod}}^k$  for a step-up stimulus and  $C_i^k, i = 0$  to  $8, C_{\text{mod}}^k, C_{\text{neg}}^k, C_{\text{pos}}^k, T_{\text{neg}}^k, T_{\text{pos}}^k, z^k$  for an impulse stimulus), the problem involves seven flow rates  $F_i$ , eight volumes  $V_i$  and 10 binary variables  $y_i$  related to the eight reactors, the by-pass and the recycle.

The objective function is given by

$$f = \rho \times \sum_{k=1}^{k_{\text{max}}} (C_{\text{mod}}^k - C_{\text{exp}}^k)^2 + \sum_{i=1}^{10} y_i \quad (10)$$

where  $\rho$  is a positive coefficient introduced to balance the orders of magnitude of the two terms of the sum. From this objective function, the goal is to obtain the simplest model (by minimizing the sum of  $y_i$ , giving also a concentration as close as possible of the experimental concentration by minimizing  $\sum_{k=1}^{k_{\text{max}}} (C_{\text{mod}}^k - C_{\text{exp}}^k)^2$ ). After numerous trials, the value 1000 has been assigned to  $\rho$  for realizing a trade-off between the two terms of the objective function.

The applicability of the optimal model defined by Eqs. (1)–(10) is quite general. It can be used for any ventilated enclosure of volume  $V_{\text{Total}}$ , but the results strongly depend on the volume value and the shape of the RTD curve.

### 3. Numerical solution

In the previous works of Montastruc et al. (2004) and Laquerbe, Laborde, Soares, Floquet, et al. (2001) and Laquerbe, Laborde, Soares, Ricciardi, et al. (2001), a two-step procedure combining a stochastic procedure (simulated annealing or genetic algorithm) for building models, and a NLP algorithm for optimizing the operating conditions of the elementary cells of the current model, was implemented. This approach appears to be attractive, but the handling of the NLP problem for each model structure proposed at the upper level by the stochastic method, poses non-trivial numerical problems. Indeed, the dimensions of the NLP problem may vary according to the proposed models, in terms of some variables and constraints that may disappear or come back into the problem. Furthermore, the stochastic procedures are unable to provide an optimal solution. For these reasons a general purpose MINLP solver was used in this paper. Insofar as it constitutes now a standard in process engineering, as well as in research and teaching fields, the GAMS package was chosen.

From handling MINLP problems, GAMS (2004) proposes two codes for solving the master problem. DICOPT++, based on the OA/ER procedure, first proposed by Duran and Grossmann (1986) and Kocis and Grossmann (1987), and improved by Kocis and Grossmann (1988) for partially non-convex problems is the oldest MINLP procedure of GAMS. Simple Branch and Bound (SBB) is the other code implemented more recently in the GAMS library. It is based on an implicit enumeration procedure first proposed by Gupta (1980) and Gupta and Ravindram (1981). At each node of the tree representing the mixed-integer linear programming (MILP) problem, corresponding to the master problem in the general MINLP solution procedure, a continuous NLP problem obtained by assimilating some discrete variables to continuous ones, is solved. This solution provides a lower bound value on the objective function, used for defining the branching scheme (two options are provided - depth first or breadth first branching) or to cut a branch (Floudas, 1995). The pseudo-cost method can be used for the selection of the next variable or the next node to branch. However the pseudo-cost computation, based on a measure of the NLP unfeasibility may require high computational times (Broke, Kendrick, Meeraus, & Raman, 1998). In the most classical version of SBB with a depth first scanning procedure, the well-known backtracking method is implemented for identifying the next node to be separated. Whatever the algorithm used for solving the master problem, AO/ER or SBB, a NLP problem has to be solved at each iteration of the MILP. For this purpose, the solver used is the classical CONOPT3, based on the GRG method first proposed by Abadie (1968) and Abadie and Carpentier (1969). The GRG implementation is detailed in the paper of Drud (1985).

In the thesis of Hocine (2006), the two solvers have been compared on the basis of various types of benchmark problems found in the literature (Floquet, Pibouleau, & Domenech, 1989; Himmelblau

**Table 1**  
Reference example for a step-up stimulus

Reference model	Solution
Case 1: two reactors in series of volume 50	Idem, QD = 0
Case 2: two reactors in parallel of volumes 10 and 15, flow rates = 2 on the two branches	Idem, QD = $3.2 \times 10^{-10}$
Fig. 2a	Fig. 2b, QD = $1.3 \times 10^{-8}$
Fig. 3a	Fig. 3b, QD = $8 \times 10^{-5}$

& Edgar, 1986). It follows from this study that SBB seems to be less influenced by the variable initialization than DICOPT++, so it has been retained for solving the problem considered in this paper.

#### 4. Validation on theoretical examples

In this section, the problem consists in trying to find again a given solution from the superstructure shown in Fig. 1. The dimensionless volumes and flow rates of this reference solution were fixed arbitrarily, the RTD curves were simulated in an Excel file, the value one is assigned to the step time  $h$ , and the time interval is [0,50].

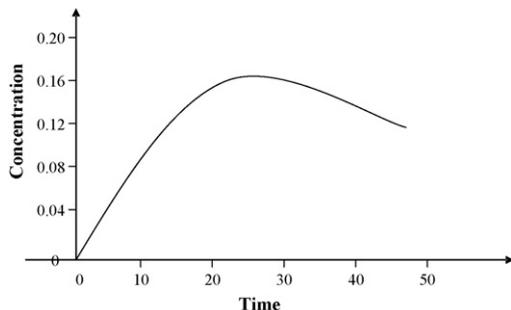
For a step-up stimulus (respectively for an impulse stimulus), the reference model is given in cases listed in the first column of Table 1 (respectively of Table 2), and the corresponding solutions found by GAMS are reported in cases listed in the second column. In these Tables, the term  $QD = \sum_{k=1}^k \max(C_{mod}^k - C_{exp}^k)^2$  represents the total quadratic deviation between experimental and modelled points. For illustrative sake, the RTD curves corresponding to an impulse stimulus and models for cases 1 and 2 of Tables 1 and 2 are respectively reported in Figs. 4 and 5, where the experimental and modelled responses cannot be distinguished.

This preliminary study shows that good solutions, with a very slight difference between the experimental and modelled RTD curves, can be extracted from the superstructure of Fig. 1 by means of the SBB GAMS's solver. In most cases, the modelled solutions are identical to the reference, and when they differ, because the GAMS solver has reached a different local optimum, the quadratic deviation is always very low.

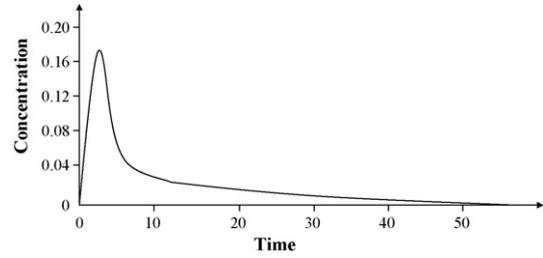
For the superstructure of Fig. 1 the previous study shows that GAMS gives good results, but these solutions probably depend to a certain extent on the superstructure. The study of the superstructure

**Table 2**  
Reference example for an impulse stimulus

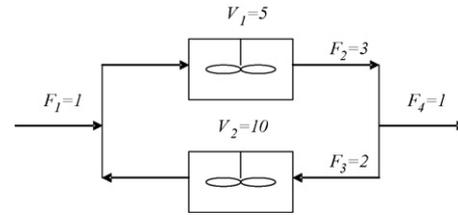
Reference model	Solution
Case 1	Idem, QD = $4.6 \times 10^{-10}$
Case 2	Idem, QD = $4.2 \times 10^{-10}$
Fig. 2a	Idem, QD = $2.9 \times 10^{-12}$
Fig. 3a	Idem, QD = $1.9 \times 10^{-7}$



**Fig. 4.** RTD curve for an impulse stimulus and model of case 1.



**Fig. 5.** RTD curve for an impulse stimulus and model of case 2.

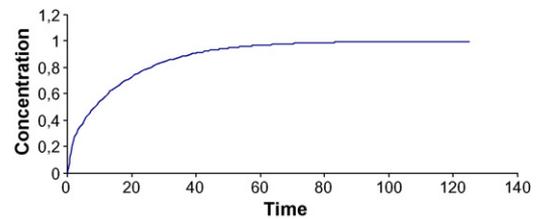


**Fig. 6.** Reference model of Ameer.

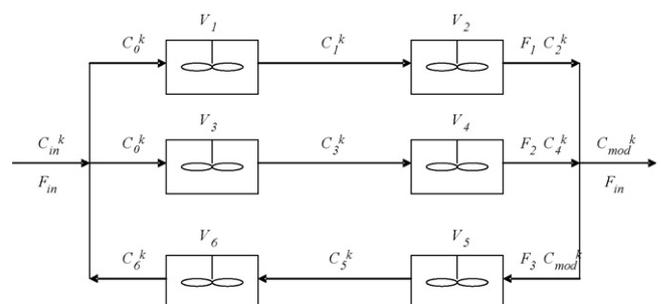
influence on the obtained solutions is now carried out through a problem proposed by Ameer (1983). The reference solution is shown in Fig. 6. Ameer (1983) has shown that the response of the reactor network of Fig. 6 to a step-up stimulus was given by the following equation:

$$C_{exp}(t) = [1 - 0.21113 \exp(-0.74641t) - 0.78867 \exp(-0.05359t)] \quad (11)$$

The experience was carried out in the time interval [0,125] min, the corresponding RTD curve is plotted in Fig. 7. In addition to the superstructure of Fig. 1, another superstructure shown in Fig. 8 was studied. Insofar as the superstructure of Fig. 8 does not involve any PFR, one can think that this type of superstructure would be more adequate for modelling the Ameer's example. The results obtained with the two superstructures and for 10 and 250 discretization points in the time space are reported in Table 3. It follows from this



**Fig. 7.** RTD curve for the Ameer's example (step-up stimulus).



**Fig. 8.** Superstructure for Ameer's example.

**Table 3**  
Results for the Ameer's problem

Number of discretization points	Superstructure of Fig. 1	Superstructure of Fig. 10
10	Fig. 9, QD = $5.6 \times 10^{-11}$	Same as Fig. 9 with $V_1 = 6.14, F_2 = 1.63,$ $F_3 = 0.63, V_2 = 8.26$ QD = $5.610^{-11}$
250	Fig. 10, QD = $2 \times 10^{-9}$	Fig. 10, QD = $2 \times 10^{-9}$

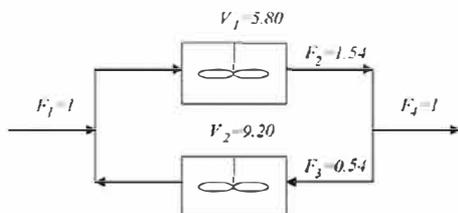


Fig. 9. Solution for the Ameer's example with the superstructure of Fig. 1 and 10 points of discretization.

study that for a given number of discretization points, the superstructure has no influence on the topology of the solution found, the only differences between the solutions lies in the values of the operating conditions. Indeed, different superstructures may lead to different local optima with the same reactor network. In opposition to the superstructure, the number of discretization points seems to have a significant effect on the solution topology. For example, with 10 points the obtained topology (Fig. 9) is the same as the reference (Fig. 6), even though with 250 points, the topologies (Figs. 6 and 10) are different. In Fig. 10 the lower branch is in the opposite direction than the lower branches of Figs. 6 and 9. This behaviour may be explained by possible several local numerical solutions that can be obtained with the SBB solver. According to the discretization scheme of the CSTR (Eq. (9b)), the path leading to a solution may be different.

## 5. Application to a ventilated laboratory enclosure: MELANIE facility

### 5.1. Experimental conditions

The ventilation system is made up of blowing fans and exhaust fans, leaks or infiltrations can occur, the enclosure being in depression or in overpressure with adjacent spaces. The contaminant transfers after an accidental pollutant release or in standard working conditions can be evaluated through an adequate flow model. This allows to predict the amount of contaminant that workers can breathe, to predict also the concentrations recorded by the contaminant monitors located somewhere in the indoor space, and then to validate the pertinence of the measures given by the monitors.

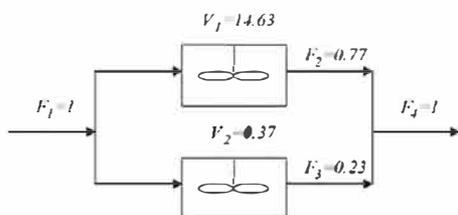


Fig. 10. Solution for the Ameer's example with the superstructure of Fig. 1 and 10 and 250 points of discretization.

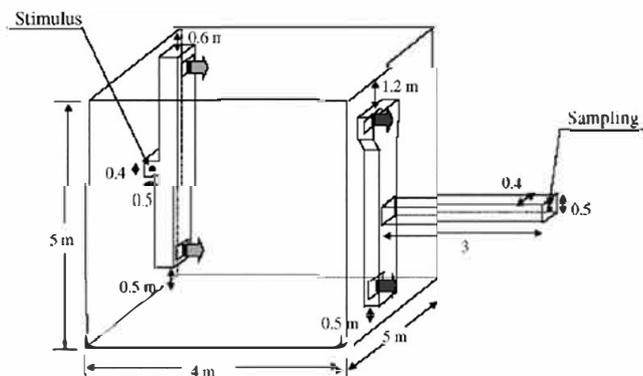


Fig. 11. MELANIE laboratory facility.

A real ventilated room of volume  $100 \text{ m}^3$ , the laboratory enclosure of the CEA (Grenoble, France), called MELANIE is now used to illustrate the application of the superstructure-based solution procedure for modelling a ventilation system. The room is shown in Fig. 11, and corresponds to an exhaust flow rate equal to  $1100 \text{ m}^3/\text{h}$  at  $20^\circ\text{C}$ , that is to say  $0.305 \text{ m}^3/\text{s}$ , and to the classical location of blowing and exhaust openings found in the nuclear applications: blowing in the upper part of the room and exhaust in the lower part. However inside this room, it is possible to obtain various air flows and residence time distributions by either changing the positions of the blowing and exhaust openings, or modifying the exhaust flow rate (Espí, 2000).

The experimental conditions correspond to a helium release used as a tracer gas, with the blowing and exhaust opening kept in open position. In this section, only the step-up stimulus is studied, because on the lack of experimental conditions concerning an impulse stimulus (tracer quantity, duration of the tracer release), which are essential to carry out a pertinent simulation. The experimental curve, obtained on a time horizon of 900s, is shown in Fig. 12.

### 5.2. Numerical studies

In all the numerical studies carried out in this section, the mean quadratic deviation (total quadratic deviation divided by the number of discretization points) is always in the order of magnitude of  $10^{-5}$ , and it is not reported on the various solutions presented.

For the superstructure shown in Fig. 1, the MINLP dimension in terms both of variables and constraints is quite important, and depends on the number of discretization points in the time space, as reported in Table 4. More than the number of variables, the presence of numerous bilinear constraints affects the convergence because it introduces non-convexities in the problem formulation and may penalize the convergence. The solution obtained for the two discretization schemes (45 and 180 points) are indicated in

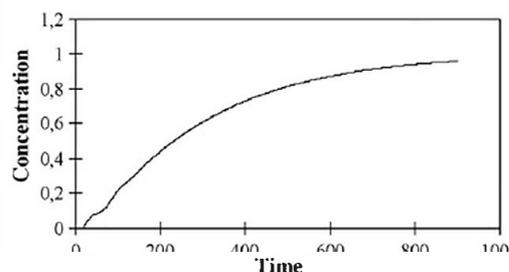
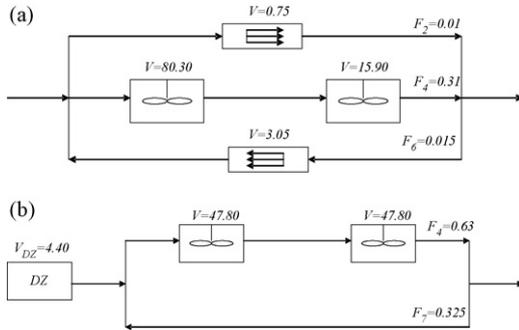


Fig. 12. Experimental response of the MELANIE room (step-up stimulus).

**Table 4**  
Dimensions of the MINLP for the superstructure of Fig. 1

Number of discretization points	Binary variables $y_i$	Volumes $V_i$	Flow rates $F_i$	Concentrations $C_0^k, C_1^k, \dots, C_8^k, C_{mod}^k$	Linear constraints Eqs. (1), (2a), 8 (4), 7 (5)	Bilinear constraints Eqs. (2b), (3), 2 (7c), 6 (9b)
45	10	8	7	450	17	450
180	10	8	7	1800	17	1800



**Fig. 13.** (a) Results for the MELANIE enclosure with 45 points and superstructure of Fig. 1. (b) Results for the MELANIE enclosure with 180 points and superstructure of Fig. 1.

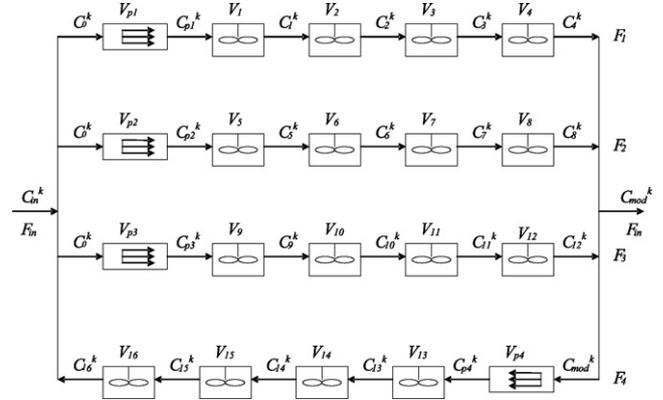
Fig. 13a and b, respectively. In Fig. 13b corresponding to the solution obtained with 180 discretization points, there is a deficiency of volume of 4.40 m<sup>3</sup>, which is interpreted as a dead zone. As in the previous example, the solutions provided by 45 and 180 discretization points are structurally different, but they are equivalent in terms of results accuracy. In both cases, the modelled points are superposed on the curve of Fig. 12. For the step-up stimulus considered here, the equivalence of results provided by the two discretization schemes in terms of accuracy of the mean quadratic deviation can be explained by the smooth (non stiff) shape of the output concentration of a CSTR expressed as a function of its input concentration (Eq. (9b)).

In order to try to improve the solution, two other studies about the superstructure influence are now presented. From the superstructure of Fig. 1, a solution involving at least a CSTR and a PFR on the same branch cannot be obtained. So, in addition to the superstructure reported in Fig. 1, two other schemes are presented in Figs. 14 and 15. The superstructure of Fig. 14 is inspired by the one proposed by Brienne, Montastruc, Martinov, and Nikov (2006) for solving a similar problem. However from the previous study of Hocine (2006) that showed that the branch proliferation does not provide any improvement on the solution, only four branches have been considered for this superstructure. The last superstructure of Fig. 15 has been built from the one of Fig. 14. As it can be noted in Tables 5 and 6, the dimensions of the MINLP problems for superstructures of Figs. 14 and 15 are more important than the dimension of the MINLP problem corresponding with the superstructure of Fig. 1. The ratios of dimensions are in the order of magnitude of about two.

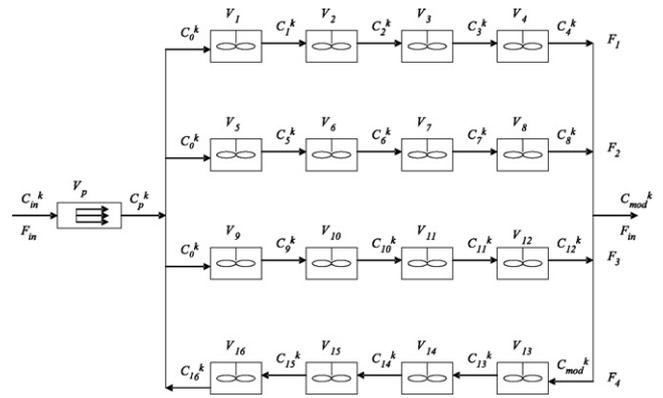
About the results obtained with these two superstructures and with two discretization schemes, contrary to the Ameur's example, both superstructures and number of discretization points have an influence on the solution reached by the solver SBB of GAMS. The

**Table 5**  
Dimensions of the MINLP for the superstructure of Fig. 14

Number of discretization points	Binary variables $y_i$	Volumes $V_i$	Flow rates $F_i$	Concentrations $C_{p1}^k, \dots, C_{p4}^k, C_0^k, C_1^k, \dots, C_{16}^k, C_{mod}^k$	Linear constraints Eqs. (1), (2a), 20 (4), 4 (5)	Bilinear constraints Eqs. (2b), (3), 4 (7c), 16 (9b)
45	20	20	4	990	26	990
180	20	20	4	3960	26	3960



**Fig. 14.** Superstructure 2.



**Fig. 15.** Superstructure 3.

accuracy of results is always in the same range, that is to say that the mean quadratic deviation is in the order of magnitude of 10<sup>-5</sup> for all the solutions presented. Although the sizes of MINLP problems are more important for superstructures of Figs. 14 and 15 than the one obtained with superstructure of Fig. 1, these new superstructures do not provide any gain with respect to the mean quadratic deviation.

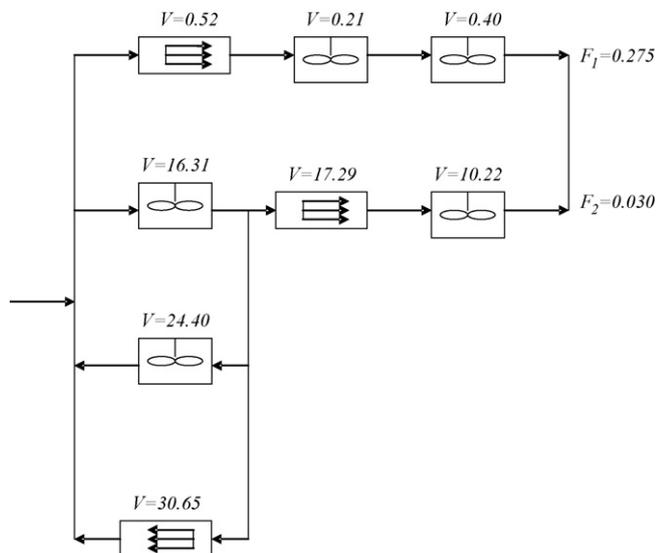
Concerning the CPU times obtained on a classical PC, they vary for one minute for the simplest case (superstructure of Fig. 1, 45 discretization points) to 2800 min for the most complex case (superstructure of Fig. 14, 180 discretization points). The CPU time grows polynomially versus the problem complexity expressed in terms of number of variables and constraints.

The solution obtained by Laquerbe, Laborde, Soares, Ricciardi, et al. (2001) by implementing a step by step strategy by merging

**Table 6**

Dimensions of the MINLP for the superstructure of Fig. 15

Number of discretization points	Binary variables $y_i$	Volumes $V_i$	Flow rates $F_i$	Concentrations $C_p^k, C_0^k, C_1^k, \dots, C_{16}^k, C_{mod}^k$	Linear constraints Eqs. (1), (2a), 17 (4), 4 (5)	Bilinear constraints Eqs. (2b), (3), (7c), 16 (9b)
45	17	17	4	855	23	855
180	17	17	4	3420	23	3420

**Fig. 16.** Solution of Laquerbe, Laborde, Soares, Ricciardi, et al. (2001).

elementary patterns, is recalled in Fig. 16. The numerical procedure of Laquerbe, Laborde, Soares, Ricciardi, et al. (2001) combines a genetic algorithm at the upper level with a SQP at the lower one. Due to the stochastic nature of the genetic algorithm, the solution optimality cannot be established for the Laquerbe's solution. The solutions obtained in this study by using the GAMS package are structurally simpler in terms of number of elementary reactors and number of streams than the solution reported by Laquerbe, Laborde, Soares, Ricciardi, et al. (2001). In terms of solution accuracy, that is to say the mean quadratic deviation between modelled and experimental points, the solutions obtained in this study are about 10 times as better than the one of Laquerbe, Laborde, Soares, Ricciardi, et al. (2001).

## 6. Conclusion

In order to characterize flow patterns and associated transfer for ventilated enclosures, this paper presents a simple and useful tool to build residence time distribution models. For large and cluttered rooms that commonly appear in the nuclear field, the proposed solution is of great interest to prevent radiological risk. It constitutes an alternative between the experimental approach difficult to implement in real cases, and the classical CFD solution procedure which would need a very thin meshing of the enclosure to provide accurate results. Based on a superstructure were potential solutions are extracted, the method aims at fitting as well as possible an experimental residence time distribution curve. The solutions, determined by implementing the GAMS package, are composed of elementary patterns (CSTR, PFR, recycles, by-passes). The GAMS environment was chosen for solving the MINLP problem because GAMS constitutes now a standard in process engineering, as well as in research and teaching fields. The MINLP problem is described in a very simple way through a high level input language, and according to the study of Hocine (2006), the SBB solver based of a

branch and bound procedure, has been retained instead of an AO/ER strategy.

A real ventilated room, the laboratory enclosure called MELANIE, is used to illustrate the application of the superstructure-based solution procedure for modelling a ventilation system. The MINLP problem involves a lot of local optima which are all equivalent in terms of mean quadratic deviation between experimental and modelled points.

In addition to the industrial ventilation, this CAD tool will have useful applications in many chemical engineering fields which commonly use the analysis of RTD curves, especially when the model representing the physico-chemical phenomena are difficult to build by hand. For example, diagnostic and design of reactors, fluidized beds and waste water plants can be mentioned.

## References

- Abadie, J. (1968). *The GRG method for nonlinear programming*. Harvey-Greenberg: Design and Implementation of Optimization Software.
- Abadie, J., & Carpentier, J. (1969). *Generalization of the Wolfe reduced gradient method to the case of nonlinear constraints*. Optimization R. Fletcher. NY: Academic Press.
- Ameur A. B. (1983). *La ventilation d'un local nucléaire—Modélisation, validation expérimentale, conséquences*. PhD thesis, INSTN.
- Athier, G., Floquet, P., Pibouleau, L., & Domenech, S. (1997). Synthesis of optimum heat exchanger networks by simulated annealing. *AIChE Journal*, 43, 3007–3015.
- Berne, P., & Blet, V., (1998). Assessment of the systemic approach using radioactive tracers and CFD. In E. Mundt & T.G. Malmstrom (Eds.), *Proceedings of RoomVent'98*, (vol. 2, pp. 275–280).
- Brienne, J. P., Montastruc, L., Martinov, M., & Nikov, I. (2006, June 26–30). Gas maldistribution in a fermenter stirred with multiple turbines. In *Proceedings of the 6th European conference on mixing*.
- Broke, A., Kendrick, D., Meeraus, A., & Raman, R. (1998). *GAMS user's guide*. GAMS Development Corporation.
- Danckwerts, P. V. (1953). Continuous flow systems—distribution of residence time. *Chemical Engineering Science*, 2, 1–13.
- Drud, A. (1985). A GRG for large sparse dynamic nonlinear optimization problems. *Mathematical Programming*, 31, 153–191.
- Duran, M. A., & Grossmann, I. E. (1986). An outer-approximation algorithm for a class of mixed-integer nonlinear programs. *Mathematical Programming*, 36, 153–191.
- Espi, E. (2000). *Prévision des transferts de contaminants en cas d'incendie dans un local ventilé*. PhD thesis, INP Toulouse.
- Floquet, P., Pibouleau, L., & Domenech, S. (1989). Identification de modèles par une méthode d'optimisation en variables mixtes. *Entropie*, 151, 28–36.
- Floudas, C. A. (1995). *Nonlinear and mixed-integer optimization, fundamentals and applications*. Oxford University Press.
- GAMS. (2004). *GAMS the solver manuals*. GAMS Development Corporation.
- Gupta, O. K. (1980). *Branch and bound experiments in nonlinear integer programming*. PhD thesis, Purdue University.
- Gupta, O. K., & Ravindram, R. (1981). *Nonlinear mixed-integer programming and discrete optimization*. NY: Progress in Engineering Optimization, ASME., p. 27.
- Himmelblau, D. M., & Edgar, T. F. (1986). *Optimization of chemical processes*. McGraw Hill.
- Hocine, S. (2006). *Identification de modèles de procédés par programmation mixte déterministe*. PhD thesis, INP Toulouse.
- Kocis, G. R., & Grossmann, I. E. (1987). Relaxation strategy for the structural optimisation of process flowsheets. *Industrial Engineering & Chemistry Research*, 26, 1869–1880.
- Kocis, G. R., & Grossmann, I. E. (1988). Global optimisation of nonconvex mixed-integer nonlinear programming. *Industrial Engineering & Chemistry Research*, 27, 1407–1421.
- Laquerbe, C., Laborde, J. C., Soares, S., Floquet, P., Pibouleau, L., & Domenech, S. (2001). Synthesis of RTD models via stochastic procedures: simulated annealing and genetic algorithm. *Computers and Chemical Engineering*, 25, 1169–1183.
- Laquerbe, C., Laborde, J. C., Soares, S., Ricciardi, L., Floquet, P., Pibouleau, L., & Domenech, S. (2001). Computer aided synthesis of RTD models to simulate

- the air flow distribution in ventilated rooms. *Chemical Engineering Science*, 56, 5727–5738.
- Leclerc, J. P., Detrez, C., Bernard, A., & Schweich, D. (1995). DTS: un logiciel d'aide à l'élaboration de modèles d'écoulement dans les réacteurs. *Revue de l'Institut Français du Pétrole*, 50(5), 641–656.
- Levenspiel, O. (1972). *Chemical reaction engineering* (2nd ed.). Wiley-Interscience.
- Montastruc, L., Azzaro-Pantel, C., Pibouleau, L., & Domenech, S. (2004). A systemic approach for pellet reactor modeling: Application to water treatment. *AIChE Journal*, 2514–2525.
- Thereska, J. (1998). L'application des radiotraceurs dans les unités industrielles-bilan et perspectives, 1<sup>er</sup> Congrès Français Traceurs et Méthodes de Traçage, Nancy. *Récents Progrès en Génie des Procédés*, 12(61), 1–8.
- Walter, E., & Pronzato, L. (1994). *Identification de modèles paramétriques à partir de données expérimentales*. Masson.
- Yuan, X., Zhang, S., Pibouleau, L., & Domenech, S. (1988). Une méthode d'optimisation non linéaire en variables mixtes pour la conception de procédés. *Recherche Opérationnelle/Operation Research*, 22, 331–346.