

# Radioactive Tracing as Aid for Diagnosing Chemical Reactors

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**Résumé — Le traçage radioactif comme aide au diagnostic des réacteurs chimiques** — Le traçage radioactif apparaît souvent comme le seul moyen de caractérisation fine et non perturbatrice des écoulements de matière dans les réacteurs chimiques. Sous réserve de l'observation de certaines conditions méthodologiques, ce traçage permet d'étudier, au sens du génie des procédés, l'écoulement d'une phase dans un réacteur, par exemple par l'obtention des distributions des temps de séjour locales ou distributions d'âges internes. Ces distributions permettent aussi de déterminer la nature de l'écoulement tracé aux fins d'extrapolation d'échelle ; on donne l'exemple de l'étude de l'hydrodynamique d'une colonne à garnissage. Elles peuvent aussi constituer des moyens de validation des codes numériques de mécanique des fluides, comme l'illustre l'exemple du traçage d'un réacteur de cristallisation par désublimation d'un gaz chaud par un gaz froid. Dans celui-ci, l'aérodynamique numérique est confirmée par la construction expérimentale d'un modèle systémique équivalent ; la zone réactionnelle est alors localisée numériquement avec un réel degré de confiance. Une propriété spécifique du traçage radioactif réside dans le fait que le signal enregistré par une sonde nucléaire est inversement proportionnel au débit volumique du traceur au voisinage de cette sonde. On peut alors remonter à la température locale par bilan enthalpique, comme l'illustre l'exemple du traçage d'un four de trempe. Ces mesures, si elles peuvent mettre en évidence des maldistributions, ne peuvent les localiser ; c'est l'objet des techniques d'imagerie, dynamiques ou non. Parmi elles, la tomographie d'émission de photon unique (TEPU) permet de reconstruire l'image de la répartition du traceur en écoulement dans une section d'un réacteur cylindrique à partir d'un nombre limité de détecteurs nucléaires. Les avantages et limitations de cette technique sont discutés et un exemple d'application à la localisation de maldistributions est donné. Enfin, le traitement des images données par une gamma-caméra permet aussi de visualiser et d'estimer la répartition locale d'un traceur radioactif dans un réacteur catalytique triphasique de laboratoire de type Mahoney-Robinson, utilisé pour la désulfuration des gazoles. L'effet de l'agitation sur l'efficacité locale des transferts liquide et gazeux est mis en évidence.

Mots-clés : radiotraceur, interactions photon-matière, codes Monte-Carlo, CFD (simulation numérique de la mécanique des fluides), tomographie, problèmes inverses, simulation.

**Abstract — Radioactive Tracing as Aid for Diagnosing Chemical Reactors** — Radioactive tracing is often the only fine and non-intrusive technique for characterising the flow of phases in chemical reactors. Provided that methodological constraints have been respected, this tracing allows to study, from a chemical engineering point of view, the flow of a phase by the measurement of local residence-time distributions (RTD) or internal ages distributions. From these distributions one can deduce the nature of the marked flow, which is required for scale extrapolation; the example of a packed column is given to illustrate this point. These distributions may validate computational fluid dynamics (CFD) codes as it is pointed out in the tracing of a crystallisation process by desublimation of a hot gas by a cold one.

*In this case, computed aerodynamics is confirmed by the experimental build-up of a systemic model; the reaction zone is then numerically localised with a high degree of confidence. A specific property of the radioactive tracing is that the signal monitored by a nuclear probe is inversely proportional to the volumetric flow rate near the probe. Then, modelling the enthalpic balance leads in a simple way to the local corresponding temperature as it is featured in the example of the tracing of a quenching tower. These measurements, even if they can point out maldistributions of flows, are not suitable for their localisations; this is the purpose of static or dynamical imaging techniques. Among them, the single photon emission computed tomography (SPECT) enables to reconstruct the image of the repartition of a tracer in the cross-section of a cylindrical reactor from a limited number of nuclear detectors. Advantages and limitations of SPECT are presented and an example of localisation of maldistributions is given. Finally, the correct treatment of images given by a gamma camera constitutes an interesting way for the visualisation and estimation of the repartition of a radioactive tracer in a laboratory three-phase catalytic reactor of the Mahoney-Robinson type used for the desulfurization of gas oils. The influence of the agitation speed on the local efficiency of the gas and liquid mass transfers can be pointed out.*

*Keywords: radiotracers, photon-matter interactions, Monte-Carlo codes, CFD, tomography, inverse problems, simulation.*

## INTRODUCTION

In industrial applications radioisotopes are often used as radiotracers injected into the flow field of interest in a known manner in order to obtain the needed information about some aspects of the flow pattern. In such main applications radiotracers provide unique information about the system which is not readily obtainable by any other means. Therefore, safe, responsible and knowledgeable use of radiotracers enables to obtain information in opaque systems, which are prevalent in industrial practice. The widespread use of tracer methodology was well illustrated by the recent congress on Tracers and Tracing Methods held in France (Leclerc and Grevillot, 1998). Processing of radiotracer data often requires the use of RTD theory for the quantification of flow patterns by identification of the transfer function (*i.e.* the impulse response). However, the proper implementation of this theory in practice requires strict adherence to a protocol for tracer injection, detection and data processing. On the other hand, advanced computational power allows one to compute flow fields in complex enclosures and equipment. However, the physics of phase interaction especially in multiphase flows remains uncertain and this cannot be overcome by computational power alone. Hence, if CFD is to be used in design of multiphase units it must be experimentally verified, and again only radioisotopes are capable of providing this vital information.

The purpose of this paper is to highlight the developments and trends in the applications of such radioactive techniques in some fundamental fields in process engineering.

## 1 PHOTONIC INTERACTIONS

Since the industrial reactors are mostly opaque, the usual radiotracers are  $\gamma$ -emitters and therefore emit photons which undergo multiple random interactions (with the fluid itself,

the walls, the screens, the collimator, etc.) until they reach the detection probe. Monte-Carlo codes (Tola, 1996) have thus been developed in order to model them and first validations using point sources in static conditions of detection have already been performed with no adjustment of any parameter (Blet *et al.*, 1999a). Recently some investigations in dynamic conditions have been conducted by labelling with  $^{133}\text{Xe}$  a gas flowing in a Plexiglas tube under well controlled conditions (*Fig. 1*). The Plexiglas test section is 30 m long and 60 mm in diameter. Three detectors were positioned on this tube respectively at 153 (detector 1), 253 (detector 2) and 455 (detector 3) tube diameters from the injection point. Three experiments have been conducted with gas flow rates respectively equal to 5.2, 10 and 239  $\text{m}^3/\text{h}$ . Figure 2 illustrates a typical set of signals corresponding to the last experiment. The deconvolution of a given signal (detector 2 or 3) by the signal from the previous detector (respectively detector 1 or 2) leads to a transfer function which can be perfectly fitted by a plug flow with dispersion (PD) function as it is shown in Figure 2.

The Monte-Carlo code simulates the interactions between the photons emitted by  $^{133}\text{Xe}$  and the different materials, provided that the concentration of the tracer is homogeneous in each cross-section of the duct. Then, coupling both the above mentioned PD function and this simulation leads to a reconstructed experimental curve which is compared with the actual experimental one in Figure 3. The good agreement between both curves could still be improved by optimising the energy thresholds. As a key feature, this agreement indicates that the deconvolution of two consecutive experimental signals should avoid any possible photonic effects as it has been already shown for other systems (Blet *et al.*, 1999a). This result has to be experimentally verified for any studied medium but as far as we can assume that the photonic interactions are linear—in the systems theory sense—it should make sense.

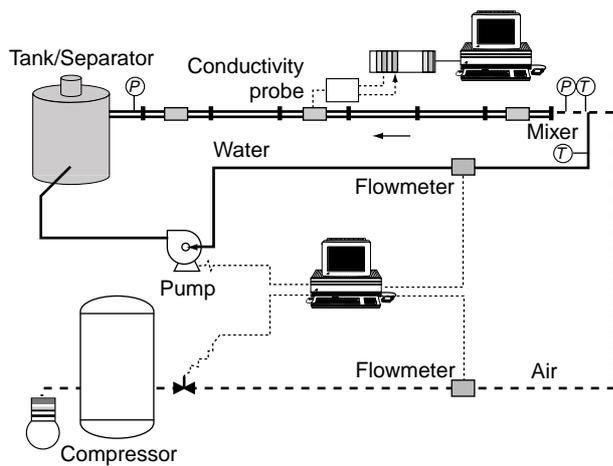


Figure 1

Experimental loop used for the measurement of phasic flow rates in air-water mixtures.

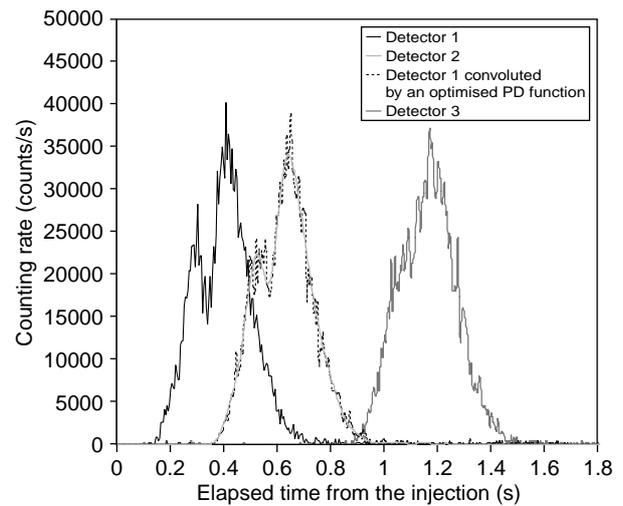


Figure 2

Typical set of experimental signals obtained from test 3 (gas flow rate = 239 m<sup>3</sup>/h). Assessment of a plug flow with dispersion (PD) as the transfer function between detectors 1 and 2.

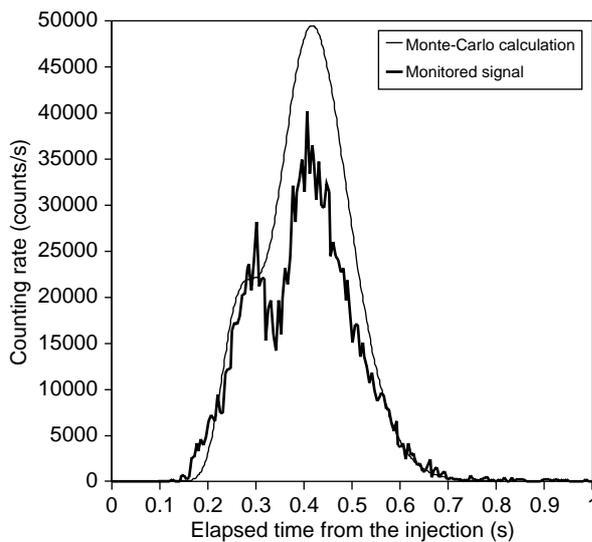


Figure 3

Comparison between the actual monitored signal corresponding to detector 2 and the one obtained by convolution of the Monte-Carlo simulation of photonic interactions with the optimised transfer function between detectors 1 and 2.

## 2 FLOW CHARACTERISATION

Flow pattern detection for air-water mixture—or slurry—flows is a complex problem of practical importance far from being exhausted (Vassalo and Kumar, 1999; Keska and Williams, 1999). First studies (Evans, Robertson and

Spackman, 1971) have already pointed out the contribution of the radiotracer technique to an understanding of the mechanics of particular two-phase flows due to its ability to make non-intrusive vapour and liquid mean velocities measurements. Thanks to the recent progress made in the modelling of photon-matter interactions and in advanced statistical data analysis techniques the question arises of the extension of this “radioactive” contribution to the phasic flow rate measurements (including velocity and void fraction measurements) in a wider range of flow conditions. From a theoretical point of view, preliminary simulations are thus being conducted in different configurations (bubbly to annular flow) and for different radiotracers either gaseous or liquid since the monitored count rate  $T$  is directly proportional to the void (or the complementary liquid) fraction  $\varepsilon$ . As a matter of fact it seems that the variation in  $T$  resulting from a variation in  $\varepsilon$  is hidden by the dispersion of the gas phase in bubbly flow.

From an experimental point of view, first results involving the above mentioned air flow tests illustrate the capability of the technique to determine the length of establishment of a flow regime (or the mixing length) but also the resulting dispersion of the flow. For example, from the previous fitted PD functions, values of the velocity  $u$  and of the coefficient of dispersion  $D$  of the air flow in the two measurement sections have been obtained. The observed variations of  $u$  and  $D$  can be attributed to an insufficient mixing of the tracer in the gas phase as it flows in front of the first detector. The fitting of the curves by a dispersive model is thus quite questionable. However, the experimental values of  $D$  have been found to be in good agreement with the theoretical ones in the highly turbulent (Taylor, 1954) and in the transition (Villermaux,

1982) zones. In the transitional flow regime the above mentioned Taylor's correlation gives a coefficient of dispersion twice lower than the measured one.

### 3 SCALE-UP PROBLEMS IN PROCESS ENGINEERING

When dealing with the interpretation of tracers experiments in multiphase contactors it is quite usual to use monodimensional continuous models like the PD function in order to extract the Peclet number of the marked phase. This has been considered for long as the best way for scale extrapolation of the reactors. Following this idea, flow characteristics of a countercurrent gas-liquid packed column were investigated using radioactive labelling of both phases (Blet *et al.*, 1999b). Velocities and coefficients of dispersion were thus obtained by the fitting of the experimental RTDs as it is depicted in Figure 4. It was found that in the vicinity of the gas distributor the gas velocity is significantly larger than in the higher parts of the column. Meanwhile, the liquid

dispersion was found to increase continuously as the fluid moves down the column but in an especially significant way near the bottom. Axial dispersion may in fact result from two causes: the truly diffusive character of the flow (due to molecular or turbulent diffusion), or the existence of a velocity profile in the radial direction (*i.e.* a "convective" effect). It is in theory possible to determine which is predominant: RTD variance should be proportional to axial distance in case of purely diffusive effects, and to the square of distance in case of convective effects. Figure 5 shows a typical plot of the second moment as a function of distance from the uppermost detectors. The resulting points seem to be correctly fitted by a parabola, but no definite trend can be given. Probably both convective and diffusive effects occur. This evolution could then be responsible for the considerable discrepancies in the liquid and gas Peclet numbers given in the literature. It also sheds some doubt on the soundness of a global input-output approach based on the dispersive plug flow model. In that case scale extrapolation is clearly hazardous.

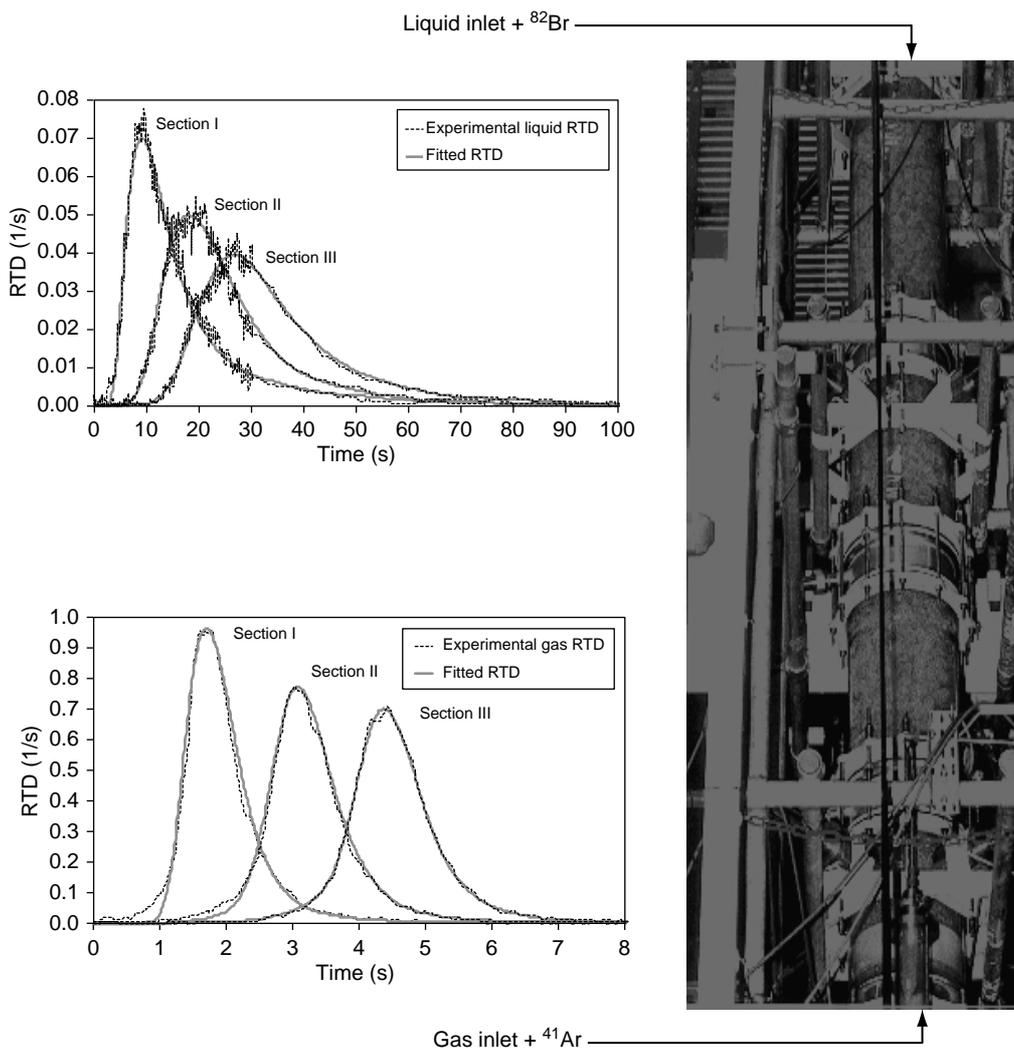


Figure 4

Gas and liquid tracing experiments in a packed bed.

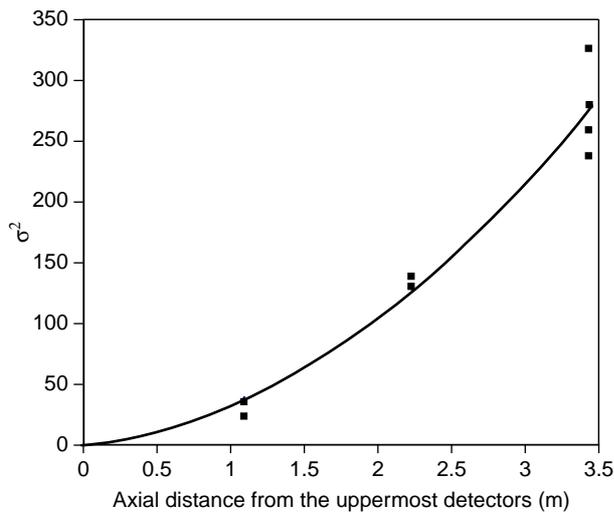


Figure 5

Evolution of the second moment as a function of the axial distance from the top of the column (water inlet).

In the same way, it is worth recalling that a radioactive measurement  $T$  (number of counts integrated during the acquisition time) is not directly related to a mixing cup or cross-sectional concentration since it is sensitive to the radial velocity profile:

$$T = \int_{\Omega} \frac{k_i C_i}{v_i} d\Omega$$

where the subscript  $i$  stands for the local value of the concentration  $C$  and the velocity  $v$  of the tracer in the elementary part  $d\Omega$  of the detection volume  $\Omega$  and  $k_i$  denotes the corresponding coefficient of calibration of the detector (*i.e.* the count rate due to the presence of the tracer uniformly distributed in the part  $d\Omega$  with a unitary concentration).

Thus, it appears that in such rather convective systems the concept of RTD cannot be applied in a “through the wall” measurement technique (Briens, Margaritis and Wild, 1995). Then the impulse response must be rather interpreted as a transfer function.

#### 4 TEMPERATURE MEASUREMENT

It is useful in particular reactors (furnaces, rotary kilns, quenching towers, etc.) to have a good idea of the temperature of the fluid flowing inside. Unfortunately, due to the extreme conditions (temperature, pressure, etc.) which can prevail in these reactors it is often quite impossible to introduce any thermometer inside the reactor. Here again, the radioactive tracing methodology can apply since the consequence of the above relationship between the count rate and the velocity is that the area  $S$  of a signal monitored by a

nuclear  $\gamma$ -probe (*i.e.* the integration of count rates during the whole residence time of the tracer and expressed as counts) is proportional to the ratio of the total activity  $A$  of this tracer divided by the volumetric flow rate  $Q$ . This relationship holds when the velocity profile can be considered as uniform in the detection volume. Thus, provided that the evolution of  $Q$  is related to the corresponding variation of temperature of the marked fluid, this property can be used with some benefit as it is illustrated in the following experiment conducted in the quenching tower of an incinerator.

The hot gas phase is introduced at the top of the tower. At a certain distance from the top, cold water is supplied to lower the mean gas temperature. The air flow has been marked by argon 41 and monitored at different locations along the tower as depicted in Figure 6. Monte-Carlo simulations have then been performed in order to evaluate the above mentioned proportionality factor for each elevation of the probes due to the fact that the nature and thickness of the walls vary along the tower as the temperature lowers.

Assuming that the gas mixture (vaporised water, the air flowing from the incinerator and the air used for the atomisation) is characterised by a unique temperature and that the vaporisation of water and the heating of the vapour are adiabatic processes, the evolution of the temperature  $T$  of this mixture can be easily obtained as a function of the mass fraction  $x$  of vaporised water (with respect to the liquid-cooling water). These assumptions are required because there is no *a priori* relationship between temperature and the mass fraction of the vapour in the gaseous mixture. However, thermal leakages can be taken into account in a global manner during the heating of the cooling water from its initial temperature to its ebullition point. Under these assumptions one can deduce from the knowledge of the local flow rates the temperature profile along the tower as it is shown in Figure 7.

#### 5 VALIDATION OF COMPUTATIONAL FLUID DYNAMICS CODES

Computational fluid dynamics frequently constitutes the only alternative for the numerical visualisation of flows in multiphase reactors especially for scale-up considerations. However, physical relevance of CFD codes may be questionable in these reactors when hydrodynamics and physico-chemical interactions are coupled. Then, it seems natural to try to validate the CFD codes using the unique potentialities of radioactive tracers. Two types of experimental validation have to be distinguished: one dealing with the laboratory scale—or pilot reactor scale—and the other with the industrial scale at which the validation can only be based on the comparison between experimental data obtained by local probes and CFD simulations. On the other hand, the first type of validation involving smaller scale reactors enables a detailed validation of CFD codes by

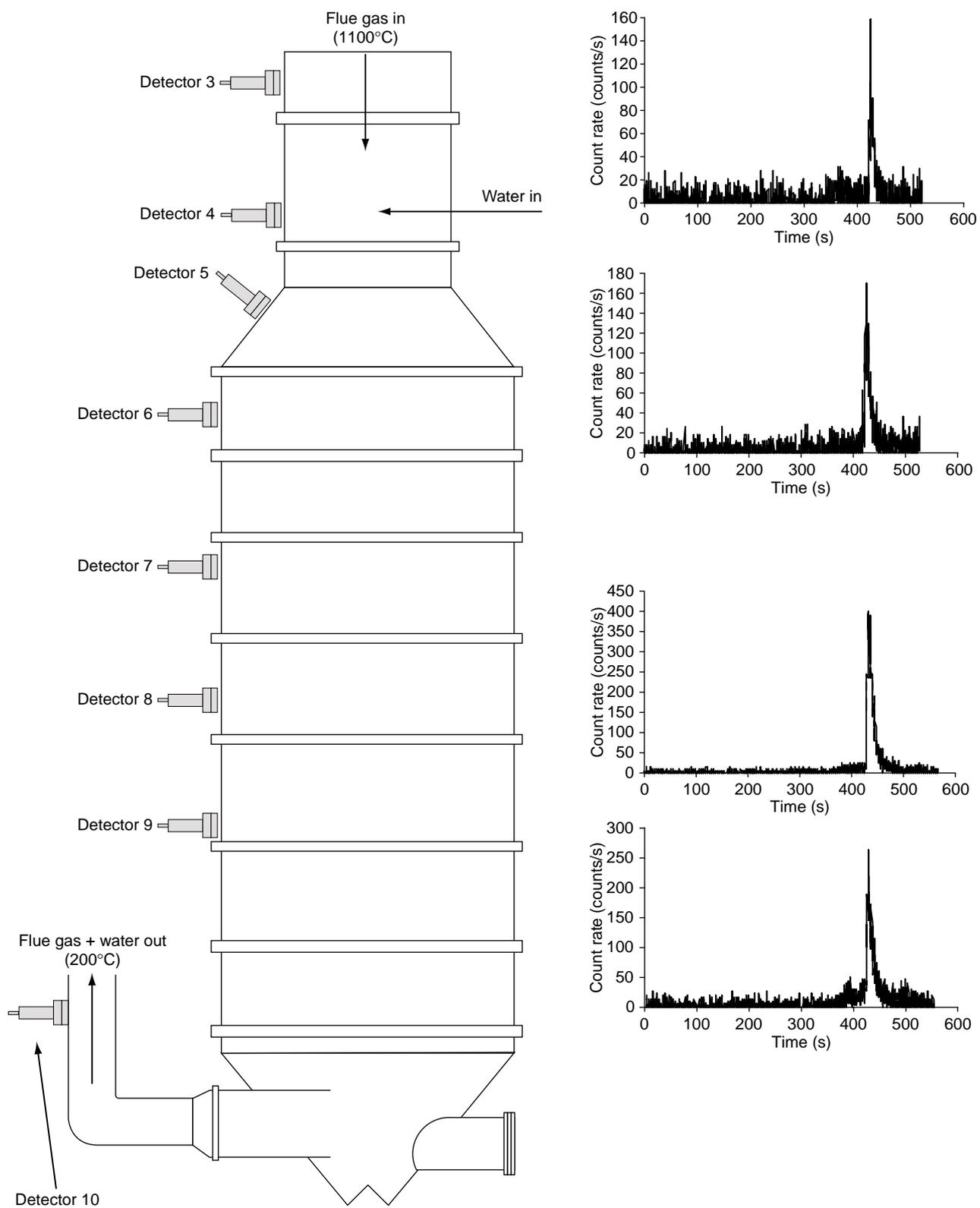


Figure 6  
Tracing of the gas phase in a waste incinerator quenching tower.

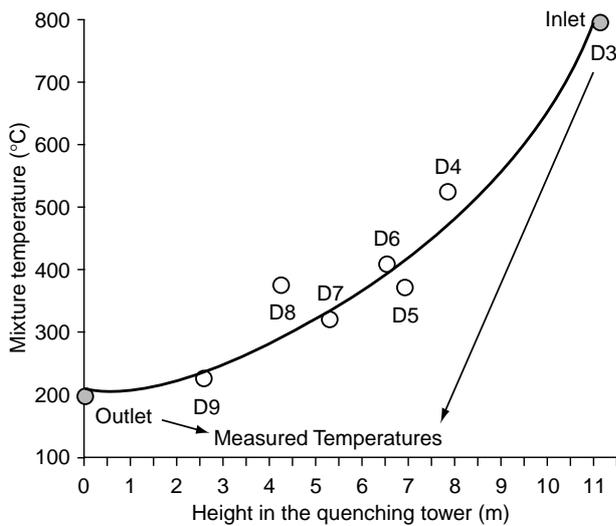


Figure 7  
Temperature profile in the quenching tower of an incinerator obtained from the enthalpic balance.

### 5.1 Industrial Scale

We had a first opportunity to make that comparison on a crystallisation reactor. This reactor is fed countercurrently by two gaseous streams, a cold and a hot one, the interaction of which creates solid particles. Tracer experiments were made on each stream, with a number of detectors sufficient to enable us to build a detailed compartmental model which is represented in the left-hand side of Figure 8. In the right-hand side of Figure 8, we have superimposed the compartmental model for hot and cold gas streams and the computed mass velocity field. This computation has been performed by CFD calculation and is described elsewhere (Blet *et al.*, 1999a). Qualitative agreement between these representations is quite remarkable, probably due to the quite flat velocity profiles. At this stage, due to the observed relevance of the CFD model the field of crystallisation mass flow rate (expressed as  $\text{kg}/\text{m}^3/\text{s}$ ) has been computed. As a surprising feature, this calculation reveals in Figure 9 that the reaction zone is very narrow and located near the inlet of the cold gas. This CFD result matches the tracing experiment from which it appeared that only a small part of the reactor was effectively used.

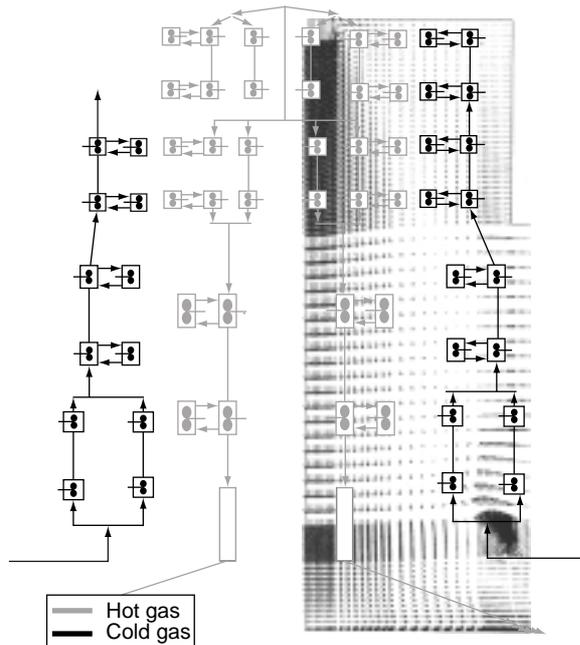


Figure 8  
Comparison between a CFD model and the compartmental approach.

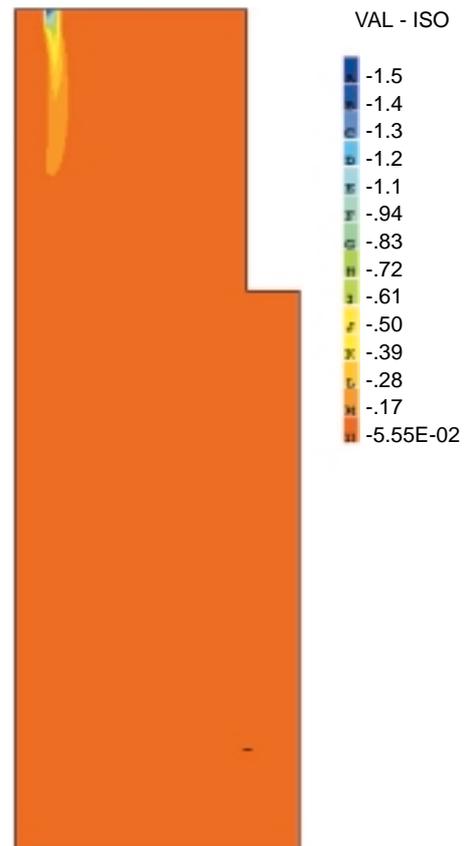
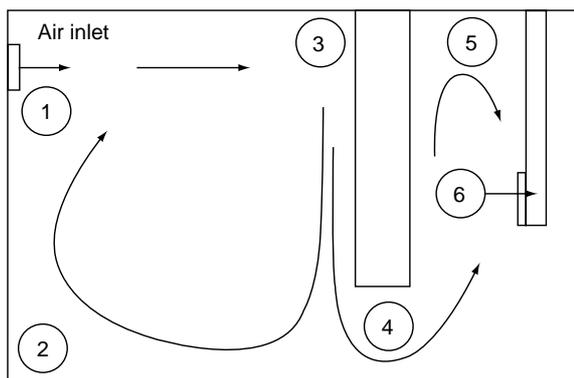
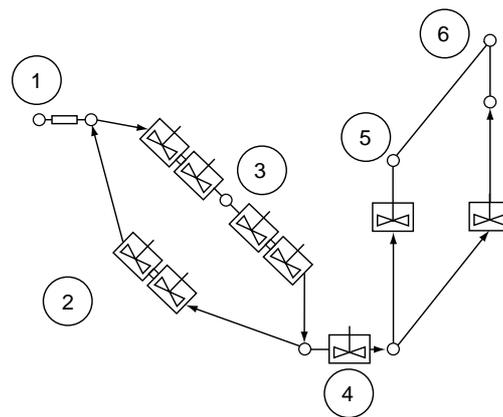


Figure 9  
Field of crystallisation mass flow rate.

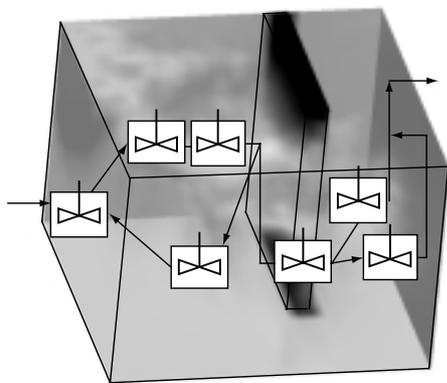
measuring the concentration and velocity fields in any cross-section of them. An extensive review of the experimental techniques dedicated to these measurements has been given by Chaouki and coworkers (Chaouki, Larachi and Dudukovic, 1997).



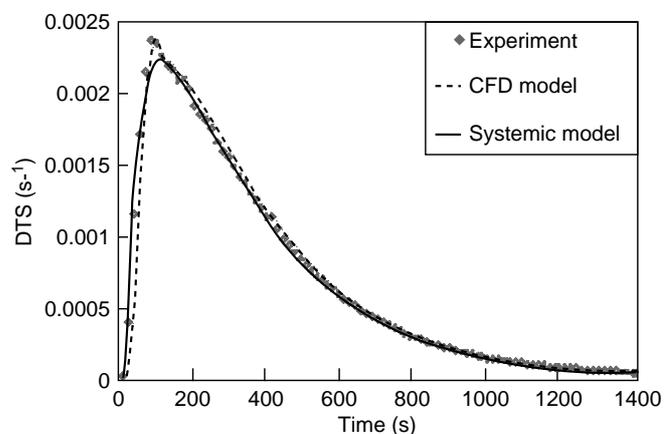
Room layout and flow pattern



Systemic model



Concentration field and systemic model



Comparison of outlet RTDs

Figure 10

Compartmental and CFD modelling of room ventilation.

Another example of comparison between tracing experiments interpreted by the compartmental approach and CFD modelling has been given in the ventilation domain. It also has been proved (*Fig. 10*) that both approaches matched not only in a qualitative description of the air flow patterns but also in a quantitative fitting of the global RTD (Berne and Blet, 1998).

When comparing these approaches one can be tempted to suggest that there is a basic link between both approaches. Theoretical work involving averaged field-based calculations in the detection volume of a nuclear probe should be undertaken to define:

- the nature of the experimental transfer function by taking into account the influence of the velocity field or the photonic interactions;
- the operational conditions for which the grid resolution of the CFD mapping is compatible with a measurement equivalent volume.

## 5.2 Laboratory Scale

A  $\gamma$ -camera is composed mainly of a very large crystal scintillator fitted with a multiple-hole collimator. Signals from a large number of photomultipliers are then processed to give a two-dimensional image of the distribution of detected photons inside the crystal. One problem is that this image bears no simple relationship with tracer concentration. If the Compton interaction can be neglected, that is to say if tracer energy is small enough, the image can be seen as an attenuation-weighted average of the concentration profiles in the direction perpendicular to the collimator. In other words, a large computing effort is required to make  $\gamma$ -camera images quantitative.

We nevertheless used a  $\gamma$ -camera (*Sopha Medical Vision*, DSX type) to try to quantify local gas hold-up as a function of impeller velocity in a Mahoney-Robinson type reactor (Blet *et al.*, 1998). Experiments were made with a low-energy

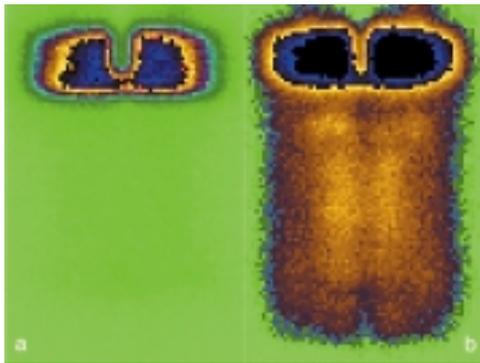


Figure 11  
 $\gamma$ -camera image of a three-phase catalytic reactor of the Mahoney-Robinson type. (a) Impeller at rest. (b) Impeller at nominal velocity.

(81 keV) gaseous tracer,  $^{133}\text{Xe}$ . Figure 11 shows two side views, with the impeller at rest (Fig. 11a) and at nominal velocity (Fig. 11b). The spot on top of the figures is the image of the gaseous ceiling. Although differences are not dramatic, gas entrapment into the liquid phase is clearly visible; an excess of gas can even be seen inside the catalyst basket, in the middle of the reactor (Figs. 12a and 12b).

### 5.3 Pilot Scale: Principles and Performances of the Detection Technique

In contrast to transmission-based techniques, the purpose of the Single Photon Emission Computed Tomography (SPECT) is to obtain transient bidimensional repartition of any marked phase—even with low contrast—in the cross-section of a cylindrical reactor (Legoupil *et al.*, 1999). In order to

reconstruct such distributions from limited view-angle sinograms, the use of algorithms such as Estimation-Maximisation (EM) requires an accurate description of the data acquisition process. Let  $p_{km}$  denote the measured projections at the  $m$ -th angle and the  $k$ -th detector. Let  $x_{ij}$  denote the unknown intensity in the pixel  $(i, j)$  and the fraction of  $x_{ij}$  that is collected in the  $p_{km}$  measurement. Then  $p_{km} = F_{ij}^{km} x_{ij} + e_{km}$  where  $e_{km}$  is the uncertainty (noise) associated to the measurement of  $p_{km}$  (Poisson probability function). The basic problem of reconstruction in SPECT consists in the inversion of the following matrix equation  $p = Fx + e$ . An element of the matrix  $F$  ( $F_{ij}^{km}$ ) is an estimator of the probability that a photon emitted from an elementary volume  $j$  of the distribution  $x$  is detected in the projection  $i$ .

The quantitative potential of SPECT relies mainly on the quality of the model (*i.e.* the estimation of the transport matrix  $F$ ) which should take into account the geometrical system response, Compton scattering, pair production, attenuation in the object and the detector responses (efficiency and collimator penetration). For that purpose, it has been shown that  $F$  is well estimated by Monte-Carlo simulations (Legoupil *et al.*, 1996a). For one detector, 110 line sources located in the plane of detectors are simulated. Each line source is estimated from 9 samples along the pipe axis in the volume of the SPECT device (Figs. 13). In order to maintain the maximum errors on probability values below 0.5%, the calculation of 990 point sources takes 10 hours on PC 200 MHz even if an acceleration algorithm based on reduction of variance technique has been implemented for the estimation of  $F$ . Due to symmetries, this calculation must be repeated three times for all the detectors. In practice, this relatively long procedure

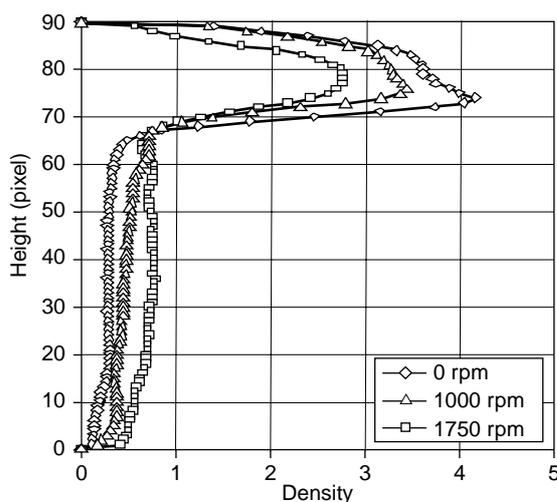


Figure 12a  
 Profile view. Decrease of the intensity of gaseous ceiling and increase of the intensity of the liquid zone as the speed of agitation increases.

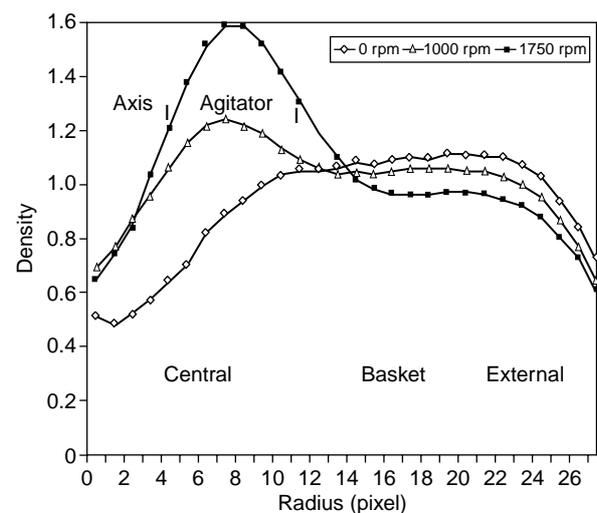


Figure 12b  
 View from the back. Increase of the intensity of the central zone and low decrease of the intensity in the catalyst basket and in the external zone as the speed of agitation increases.

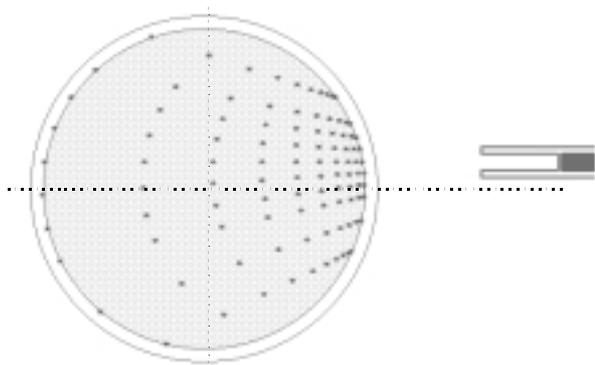


Figure 13a  
Point source locations for the estimation of the transport matrix.

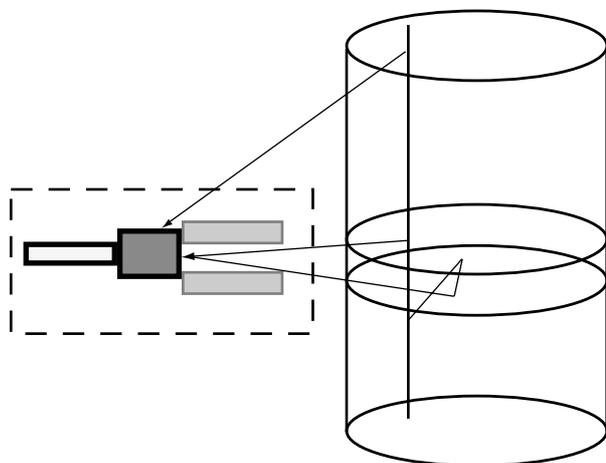


Figure 13b  
Line spread function for the estimation of the transport matrix.

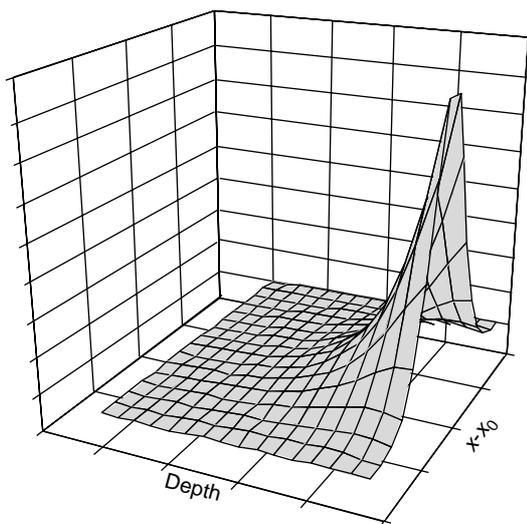


Figure 13c  
Representation of the transport matrix for one detector.

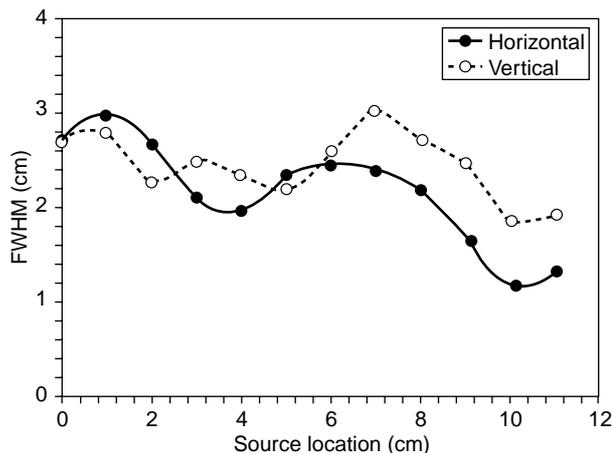


Figure 14a  
Horizontal and vertical FWHM (Full Width at Half Maximum) on a radius in the object (diameter = 24 cm) perpendicular to a projection (incidence angle = 0°).

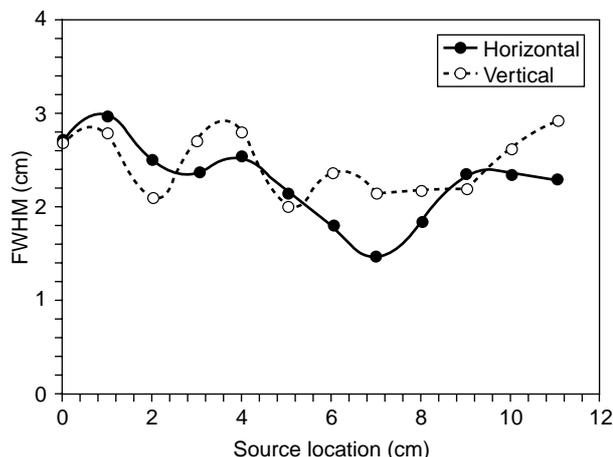


Figure 14b  
Horizontal and vertical FWHM on a radius in the object (diameter = 24 cm) bisectrical to two consecutive projections (incidence angle = 30°).

is applied once the final configuration of detectors (energy threshold, collimator and positions) is defined.

Due to the fact that the above inversion problem is an ill-conditioned one, methods of reconstruction based on the maximum likelihood as the EM algorithm are particularly convenient. This iterative algorithm not only ensures a good regularisation of the reconstruction problem but also allows one to introduce some *a priori* knowledge related to the nature of the searched distribution in the reconstruction process (Legoupil *et al.*, 1996b).

The development of an operational acquisition system based on SPECT has required many tests either simulated or experimental of static distributions of tracers representative

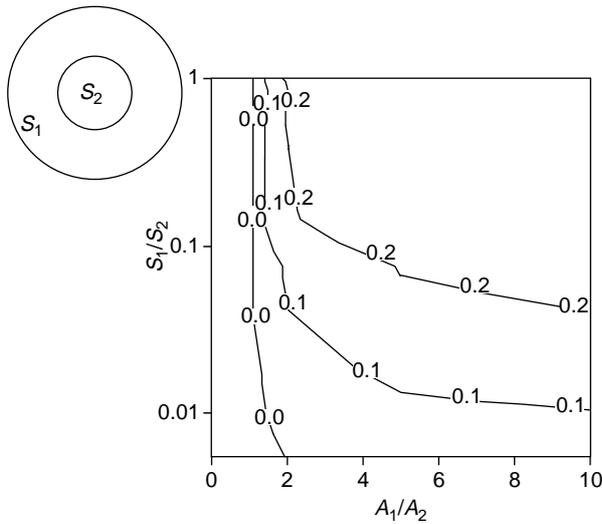


Figure 15a  
Detection properties for an annular distribution of tracer.

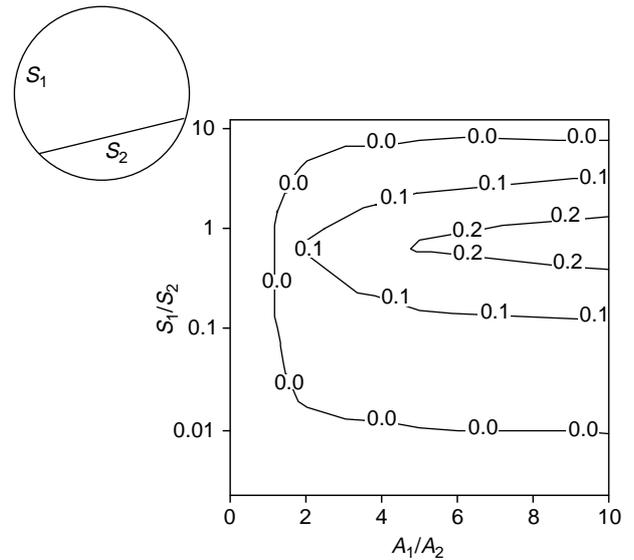


Figure 15b  
Detection properties for a stratified distribution of tracer.

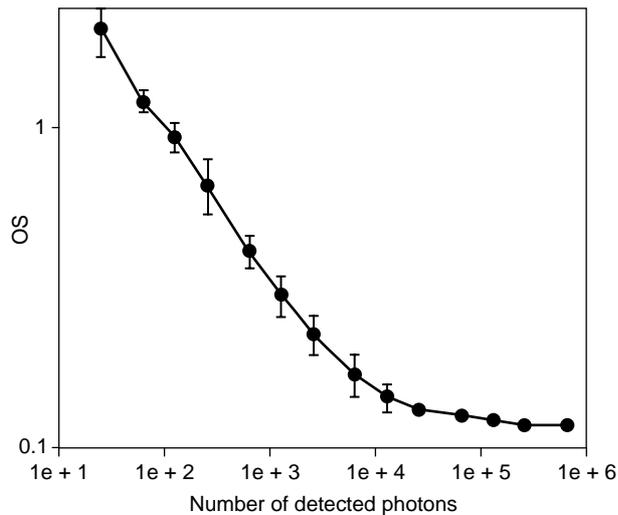


Figure 16  
Detection properties as a function of the number of detected photons.

of the diameter of the object close to the edge and 15% in the middle of the pipe as it can be shown in Figures 14.

The detection properties of the 6 x 6 detectors SPECT device have been estimated for a given tracer distribution (annular or stratified) in a cylindrical vessel. The concentration of the tracer is supposed to be homogeneous in each zone of the reactor. Numerous simulations of projection-reconstruction of the object allow to define the detection properties as a function of the surfaces ratio ( $S_1/S_2$ ) and activities ratio ( $A_1/A_2$ ) between the two zones of the studied distribution as depicted in Figures 15 which show results for an annular flow (Fig. 15a) and a stratified flow (Fig. 15b). For each couple (surface-activity), a numerical observer (OS) gives the capability of the SPECT device to identify whether or not both zones have the same statistics properties. For a given distribution the reconstructed image is all the best than the associated OS is high. The statistical noise of detection is not taken into account in this representation. Nevertheless, if it should be, it has been shown that these previous values are asymptotic to a particular function including the average number of detected photons as depicted in Figure 16.

### 5.4 Pilot Scale: Case Studies

As a validation case, the SPECT device has been applied to a 2D planar flow in a mixing vessel. The upper cover of the cylindrical vessel is made of transparent material to allow a video monitoring of the dye water tracer injected simultaneously with the radioactive tracer. The cylinder is continuously fed with a water flow rate of 0.32 l/min. Frame

of industrial configurations (Legoupil *et al.*, 1997). The best compromise (*i.e.* performances of the detection system *versus* number of detectors) has been found to consist in 36 detectors positioned in a hexagonal form assuming that spatial frequencies of phenomena in the plane of detectors we focus on are relatively low. For that solution, linearity in the reconstructed image remains acceptable under 0.1 time Nyquist frequency. Therefore, the studied object is observed under 6 sets of projections, each of them sampled over 6 measurements. The spatial resolution is about 10%

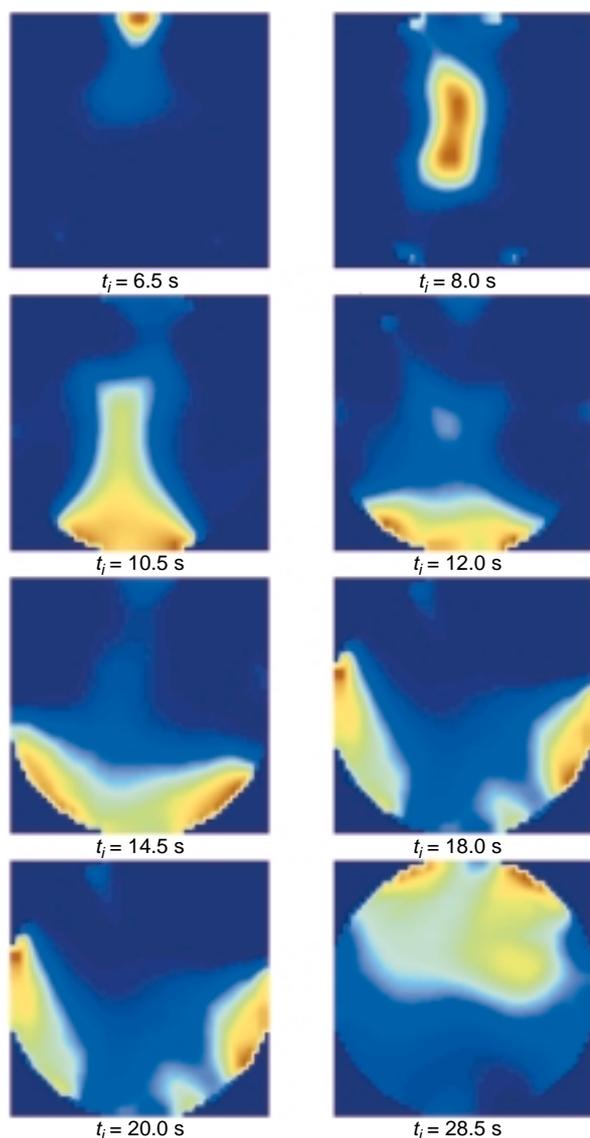


Figure 17

Reconstructed images.

acquisition time is 0.5 s. Residual air volume is insignificant and injection time is assumed to be short compared to the transit time of the tracer. Comparison of Figures 17 and 18 shows that the different sequences of the reconstructed images match in detail the video frames from which the reconstructed activities of regions of interest in the object can be obtained (Legoupil *et al.*, 1999).

Although the spatial resolution is limited by the number of detectors, one may consider that it is satisfactory for applications where statistical variations of flows are greater than this intrinsic spatial resolution of the device. Therefore, this configuration has been applied to an industrial problem where the aim of the analysis was to visualise the hot and cold water flows upstream and downstream a pipe connection. The SPECT method proved that flows

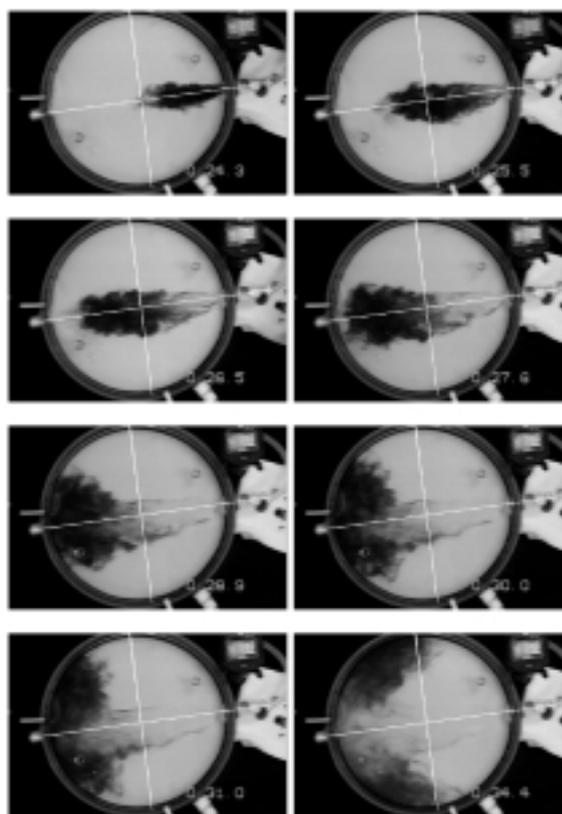


Figure 18

Video monitoring.



Figure 19

The SPECT experimental device for the detection of liquid maldistribution in a packed column.

were inhomogeneous after the connection and unexpected phenomena in the cold water inlet pipe occurred at fixed frequency.

Recently, the SPECT device has been applied for the detection of the distribution of the liquid phase in the above mentioned packed column (Fig. 19). Following the conclusions of a previous paper (Blet *et al.*, 1999b) the aim of this analysis is to localise and quantify the liquid maldistribution at the top and the bottom of the column and more especially the thickening of the liquid films at this last level (near the gas distributor).

## CONCLUSIONS

It appears clearly that the radioactive tracers have a unique role to play in the understanding of the flow patterns in opaque chemical reactors by providing tools for the validation of the computational fluid dynamics codes. However it should be emphasised that this role still requires some developments not only in the enhancement of the characteristics of imaging techniques (for example higher spatial resolution by the use of CdTe detectors) but also from a more theoretical point of view in order to refine the interpretation of data given by the above techniques. Among them, the Single Photon Emission Computed Tomography could be of great interest in the measurement of concentration and velocity fields of any studied phase flowing in these reactors.

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## REFERENCES

- Berne, Ph. and Blet, V. (1998) Assessment of the Systemic Approach using Radioactive Tracers and CFD. *Proceedings of the 6th International Conference on Air Distribution in Rooms*, Stockholm.
- Blet, V., Berne, Ph., Forissier, M., Ladet, O., Pitault, I. and Schweich, D. (1998) Apport de la  $\gamma$ -caméra à l'étude d'un réacteur agité triphasique de laboratoire. *Récents Progrès en génie des procédés*, **12**, 61, 127-133.
- Blet, V., Berne, Ph., Tola, F., Vitart, X. and Chaussy, C. (1999a) Recent Developments in Radioactive Tracers Methodology. *Applied Radiation and Isotopes*, **51**, 615-624.
- Blet, V., Berne, Ph., Chaussy, C., Perrin, S. and Schweich, D. (1999b) Characterization of a Packed Column using Radioactive Tracers. *Chem. Eng. Sci.*, **54**, 91-101.
- Briens, C.L., Margaritis, A. and Wild, G. (1995) A New Stochastic Model and Measurement Errors in Residence Time Distributions of Multiphase Reactors. *Chem. Eng. Sc.*, **50**, 279-287.
- Chaouki, J., Larachi, F. and Dudukovic, M.P. (1997) Noninvasive Tomographic and Velocimetric Monitoring of Multiphase Flows. *Ind. Eng. Chem. Res.*, **36**, 11, 4476-4503.
- Evans, G.V., Robertson, J.M. and Spackman, R. (1971) Evaluation of a Radiotracer Method of Measuring Mean Liquid Phase Velocities in Air-Water, Two Phase Flow. *Report AERE R 6753*, Harwell, Berkshire.
- Keska, J.K. and Williams, B.E. (1999) Experimental Comparison of Flow Pattern Detection Techniques for Air-Water Mixture Flow. *Experimental Thermal and Fluid Science*, **19**, 1-12.
- Leclerc, J.P. and Grevillot, G. (eds.) (1998) Traceurs et méthodes de traçage. *Récents Progrès en génie des procédés*, **12**, SFGP Ed.
- Legoupil, S. (1999) Tomographie d'émission gamma à partir d'un nombre limité de détecteurs, appliquée à la visualisation d'écoulements. *Report CEA-R-5866*.
- Legoupil, S., Pascal, G., Chambellan, D. and Bloyet, D. (1996a) Determination of the Detection Process in an Experimental Tomograph for Industrial Fluid Flow Visualization using Radioactive Tracers. *IEEE Transactions on Nuclear Science*, **43**, 2, 751-760.
- Legoupil, S., Pascal, G., Chambellan, D. and Bloyet, D. (1996b) Single Photon Emission Computed Tomography Applied to Fluid Flow Circulation Analysis by Means of Radioactive Tracer Technique. *Proceedings of the International Conference: Inverse Problems in Engineering: Theory and Practice*, Le Croisic, France.
- Legoupil, S., Pascal, G., Chambellan, D. and Bloyet, D. (1997) An Experimental Single Photon Emission Computed Tomograph for Dynamic 2D Fluid Flow Analysis. *Journal of Applied Radiations and Isotopes - Proceedings of the 3rd Topical Meeting on Industrial Radiation and Radioisotope Measurements and Applications*, Raleigh, NC, **48**, 10-12, 1507-1514.
- Legoupil, S., Tola, F., Chambellan, D. and Vitart, X. (1999) A SPECT Device for Industrial Flows Visualization. *Italian Association of Chemical Engineering Conference Series*, Florence.
- Taylor, G. (1954) The Dispersion of Matter in Turbulent Flow through a Pipe. *Proceedings of the Royal Society of London*, **A223**, 446-468.
- Tola, F. (1996) Ecrin, code Monte-Carlo de simulation de la réponse d'un détecteur à un mélange de traceurs radioactifs uniformément distribués dans une conduite cylindrique. *Internal Report*, CEA/DTA/DAMRI/SAR/ 96-111/T40.
- Vassalo, P.F. and Kumar, R. (1999) Liquid and Gas Velocity Measurements using LDV in Air-Water Duct Flow. *Experimental Thermal and Fluid Science*, **19**, 85-92.
- Villermaux, J. (1982) *Génie de la réaction chimique*, Tec. & Doc. Lavoisier.