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RECONSTRUCTION OF RADIAL CATALYST CONCENTRATION DISTRIBUTION IN AN EXPERIMENTAL TYPE FCC RISER

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Abstract: A gamma ray tomographic reconstruction of catalyst concentration distribution in laboratory experiments is presented. The images are generated by gamma transition measurements in an experimental riser simulating flow experiments. The manual scanner is adequate for mapping radial catalyst concentration distribution under spatial resolution of 0.002 m and density resolution of 10 kg/m^3.

Keywords: gamma tomography, catalyst concentration, experimental riser.

1.INTRODUCTION

The CT- Gamma Ray Computed Tomography in process engineering actually presents a rather sophisticated technological stage. Investigations on FCC – Fluid Catalytic Cracking units and on multiphase flow in opaque reactors are increasing in continuous scientific and technological developments. Nevertheless, industrial riser tomography reveals problems that often originate from the gamma transition measurements. As it is a combination of physical measuring and mathematical reconstruction techniques a closer approach to both is desired. Following this, experimental and theoretical investigations, independent of the reactor fluid dynamics, were adopted. In addition, even the most advanced CT-Tomography, uses intermediate energy such as the Cs-137, 0.662 MeV, isotopic source, NaI scintillation detector, and the reconstruction technique is based on the Beer-Lambert equation. By means of this equation the transition measurement is a function of the gamma ray pass length. To have a distributed density over the diameter surface it is necessary to formulate the σ density as a $\sigma = f(x,y)$ function. As the measured density is a mean value, by varying source and detector positions, $\sigma = f(x,y)$ values are the entrance to a data matrix, where x_i and y_i cross points are used to calculate local density.

Fluid dynamic models require parameters as gas and solid phase hold up, and gas and solid velocities that are based on measured variables, constant values as densities, which are from reference data and parameters calculated from measurement and reference data as friction coefficient and Reynolds number. The experimental data was initially modeled to evaluate instrumental precision on the measurements of: Gas flow, solid flow and the pressure drop in riser ΔP_{riser}. To estimate measured and expected values discrete models were applied by Dantas et al., (2006).

Some Basical data are not easy for comparison, as mass attenuation coefficient from the catalyst, for example. Variation in chemical composition, as well in grain size distribution are reasons for which only a very few papers as Azzi, et al., (199), gives catalyst mass attenuation coefficient data. To evaluate the precision in density measurements, catalyst attenuation coefficient and gamma intensity are the main contributions on error propagation.

2. GAMMA RAY TOMOGRAPHY

Trying to monitor the state of art of CT - Gamma Ray Computed Tomography and foreseeing its contribution to the opaque multiphase reactors let us look at the competition. "The best known technology is CAT scanning in medicine, however process tomography instrumentation needs to be cheaper, faster and more robust." This words are at the opening Industrial Process Tomography, held in Aizu, Japan, in September 2005. Among several fields of investigation that have taken this direction, the big effort devoted to electrical capacitance tomography (ECT) and to electrical impedance tomography (EIT) methods, is remarkable.

Industrial Gamma Ray Tomography – CT, as the CAT scanning in medicine are photon count techniques whose intensity is a function of the attenuation coefficient. In both cases the data goes to mathematical reconstruction and computational algorithm to generate tomographic images. Along the way, gamma ray tomography has incorporated results from the fantastic development of medicine tomography. At the actual stage, industrial Gamma Ray Tomography faces quite specific technological aspects, probably due to them, search for cheaper, faster and more robust instrumentation are on the way. In a different approach from the medicine CAT, experimental set ups in process engineering are often design, manufactured and installed by the research groups themselves since desired tomographic systems are still not commercially available. Some of these set ups are rather simple and include technological innovations such as dual gamma set ups with two ^{137}Cs gamma radiation sources (0.662 MeV) and two BGO detectors (Mudde et al., 1999). A more sophisticated self-made tomographic set up is described by Dudukovic (2005), which includes a collimated Cs-137 source positioned opposite eleven 2" sodium iodine scintillation detectors in a fan beam arrangement. In some research institutes the building of tomographic systems and the research works that are applied, are developed simultaneously, as presented by Behling and Mewes (2004). A very sophisticated and recent instrumental development is given by the Forschungszentrum Russendorf (2005), before 2005 they work with a gamma tomography detector that consists of 64 BGO scintillation crystals coupled to photomultipliers and counting electronics. The demand for higher resolution leads to the development of a high resolution tomographic gamma ray detector with 320 single detectors of 2mm x 8mm active area based on special scintillations detectors coupled to avalanche photo diodes. The system is in an arc-like array with a Cs-137 isotopic radiation source of 5 mm diameter and 180 GBq activity. This detector performs an image resolution of approximately 3mm FWHM and a structure recovery downto200µm. Competitiveness inside the CT tomography area; include industrial tomographic services such as the Tru-Tec Services, Inc. They offer the Cat-Scan technique that focuses on the hold up distribution analyses and uses contour plots that might simplify the tomographic imaging process.

Such instrumental competition certainly demands detectors and coupled electronic investigations. Again as they are photon detectors, medicine leads the demand for greater resolution and real time imaging at higher energies that is pushing the scintillation array technology. An investigation on old scintillator materials like Bismute Germanate ($Bi_4Ge_3O_{12}$), producing new BGO detectors, are being carried out. Position sensitive-light sensors have been driven to a great extent by medical radiography (Krus at al., 1999). From a more general trend or by specific demand the CT tomography for engineering process is incorporating such new instrumental achievements.

The contribution to the reactor multiphase flows study should be mentioned, at first, an impressive quotation: " It was only a decade ago that the oil industry using gamma ray techniques (Sun and Koves, 1998) learned that their large diameter risers operate in the core-annular flow regime; the core is very dilute. The core-annular structure leads to two main problems: (1) insufficient gas-solid contact, and (2) back-mixing due to non-uniform radial distributions(Jim, et. al., 1997). This unfavorable radial volume fraction distribution of solids in the riser has led to consideration of new schemes of contacting for a refinery of the 21st century ", (Gamwo, I. and Gidaspow, D. 1999). Still including radiotracer techniques as gamma ray emitter the review from (Chaouki et al., 1997a 1997b) shows that they are the most competitive techniques for investigation on large scale multiphase flow systems. More recently Dudukovic (2005) shows the advantage of combining both techniques, gamma ray computer tomography CT and computer automated radioactive particle tracking CARPT. And focuses on the main contribution from CT that is phase hold up distribution whose data are useful for validating Computational Fluid Dynamics – CDF codes. According to Dudukovic (2005) a new CT unit can achieve spatial resolution of 2 – 3 mm but requires longer scanning times of about 4 hours and yielding a density resolution better than 0.008 g/cm^3. Still a recent achievement to get better resolution at low counts by means of an Alternative Minimizing Algorithm is mentioned by (Bhusarapu et al., 2005).

The nuclear methods have the limitation of the Poisson nature of the photon generation process. High spatial resolution requires relative long measuring times as compared to the impedance techniques (Mudde, et al., (2005).

Timothy et al.(2005), describes a Gamma Densitometry Tomography (GDT) system that consists of a 100-mCi ^{137}Cs source and an array of 8 NaI(Tl) scintillation detectors. The source produces a fan-shaped beam that passes through the riser to the detector array, where the gamma intensity along each distinct ray is measured. The GDT and electrical impedance thomography EIT results show good agreement for both radial and axial solids-volume-fraction profiles. This means, that Gamma Ray CT tomography is recognized as a standard method.

3.EXPERIMENTAL

An arrangement to install, detector, gamma source and stainless steel tubes of 0. 154 m internal diameter, follows the geometry for riser irradiation. The ^{137}Cs source and detector cylindrical collimators allowed a gamma beam of 0.055 m diameter to scan along the riser radius for the transition measurements. The experimental configuration given by Azzi, et al., (1991), was implemented in a Matlab routine. The scanning interval was defined on the gamma profile to generate the density data matrix. The Compton scattering contribution was minimized by collimator length and additionally a computer program was written to improve spectrum evaluation under heavy Compton interference (Costa, et al., 2004).

Discrete models for modeling riser profile, space resolution and density resolution were implemented by computer programs. Static and simulated flow experiments were carried out to test catalyst concentration reconstruction. Density radial distributions were analyzed in 3D graphics, according to metrological criterions for data visualization, prior to the image reconstruction.

4.MATHEMATICAL RECONSTRUCTION

The objective here is to reconstruct the graph of the density function from the gamma ray trajectories mean density values. The experimental configuration used in this work assumes the trajectories are parallel to the edges of an equilateral triangle in which the riser is inscribed. Since in each of the three directions the amount of trajectories is the same, and they are uniformly spaced, this configuration induces a triangulation in this large triangle, formed only by mini congruent equilateral triangles. This arrangement favors accuracy, since any vertex in a mini-triangle lies in three trajectories, which means that the corresponding function value is present in three equations. Other three-direction-configurations will lead to irregular triangles, with a larger number of vertices, generally present in two trajectories each. Sparser areas will present larger errors than denser ones.

The novelty of this work is to present and test with experimental data, a reconstruction more suitable to this scenario of equilateral triangulation: the non-parametric Bézier triangles, to be defined in the next section. The argument for this kind of functions is a triplet of *barycentric coordinates*. If it is given in the plane three non-collinear points, it is possible to write any other point in the plane as a barycentric combination of these three points. Let **a**, **b** and **c** be three non-collinear points in the plane. Then we may say that $\mathbf{p} = u\mathbf{a} + v\mathbf{b} + w\mathbf{c}$, where $u+v+w=1$, and **p** is an arbitrary point in the plane. We say that (u,v,w) is a triplet of barycentric coordinates of **p** with respect to **a**, **b** and **c**. If **p** is in the interior of the triangle determined by **a**, **b** and **c**, then u,$v$and w take values in the interval [0,1]. The points where $u=k$, where k is a real constant, form a straight line parallel to the line segment between **b** and **c**. The points where $v=k$, where k is a real constant, form a straight line parallel to the line segment between **a** and **c**. It is analogous for $w=k$, and the segment between **a** and **b**. For instance, $w=1$, 1/3 and 0 correspond respectively to the lines passing through point **c**, the triangle's barycenter and both points **a** and **b**.

4.1 Bézier Triangles

The non-parametric Bézier triangle is a patch of a polynomial surface, in which each variable takes barycentric coordinates with respect to a given triangle in the domain. These barycentric coordinates are arguments for the Bernstein Basis, which become weights for the weighted average of the density values associated with the intersections of the gamma rays. More precisely, using the notation of [3]:

$$D(\mathbf{u}) = \sum_{|\mathbf{i}|=n} B_{\mathbf{i}}^{n}(\mathbf{u}) b_{\mathbf{i}} \qquad (1)$$

$D(\mathbf{u})$ is the proposed density function, where $\mathbf{u}=(u,v,w)$, with $u+v+w=1$ the barycentric coordinates of the point for which the density value is required, **i** is a triple-index-value ijk, used to identify each intersection point and its density value: $b_{\mathbf{i}}$. In the formula, $|\mathbf{i}|=n$ means that $i+j+k=n$. The summation runs over all possible positive integer values of ijk subject to this condition.

The Bernstein polynomials are defined as:

$$B_{\mathbf{i}}^{n}(\mathbf{u}) = \binom{n}{\mathbf{i}} u^i v^j w^k ,$$

where

$$\binom{n}{\mathbf{i}} = \frac{n!}{i!\,j!\,k!}$$

The density values $b_{\mathbf{i}}$ are called control densities, and are associated with the control points of the non-parametric triangular Bézier patch:

$$\left(\frac{i}{n} \quad \frac{j}{n} \quad \frac{k}{n} \quad b_{\mathbf{i}}\right)^t,$$

for each i,j,k so that $i+j+k=n$. The graph of D interpolates $b_{\mathbf{i}}$, where $\mathbf{i}= ijk$, where $i=n$, $j=n$ or $k=n$, the three ends of the triangle. The Bernstein polynomials form a basis for the space of all polynomials in two variables (since $u+v+w$□1 Therefore, any polynomial surface is a Bézier surface. Another important property is that they satisfy the *unit partition* condition:

$$\sum_{|\mathbf{i}|=n} B_{\mathbf{i}}^n(\mathbf{u}) \equiv 1 \qquad (2)$$

independently of $\mathbf{u}=(u,v,w)$, with $u+v+w=1$. Since

u,v,w in this present work's application are non-negative, it will always be obtained a convex combination of the control density values when computing an arbitrary point's density value. That is a desirable quality, as far as the error analysis is concerned. In the next section it is shown how the control densities are found.

4.2 Least Squares Approach

Here it is proposed to estimate the control densities through an application of the least squares method to the function defined in the equation (1). We suggest that the average of the control densities along a gamma ray trajectory is a good estimate for the actual density value from the experiments. It is not hard to realize that the bigger the number of trajectories, the better the estimation. In barycentric coordinates, the points in a given gamma ray trajectory can be characterized as those in which one of the coordinates is a fixed positive number, less than or equal to 1. According to our choices for the triangle configuration, one such a number is always of the type $i/(m-1)$ for some i in $\{0,1,...,m-1\}$, where m is the number of trajectories in each edge of the triangle that circumscribes a riser's section. We may establish that □$_{u,i}$, □$_{v,j}$ and □$_{w,k}$ are the acquired experimental density values associated with the trajectories in which $u=i/(m-1)$, $v=j/(m-1)$ and $w=k/(m-1)$, respectively. Let $\mathbf{i}/m$ be the triplet $(i/(m-1), j/(m-1), k/(m-1))$. In the trajectory where $u=i_0/(m-1)$ for some i_0, we may say that the average density of the approximating function from (1) ideally should be equal to the corresponding acquired experimental density:

$$\frac{1}{m-i_0} \sum_{\substack{|\mathbf{i}|=m-1 \\ i=i_0}} D(\mathbf{i}/m) = \delta_{u,i_0}$$

Substituting (1) into this equation we get:

$$\frac{1}{m-i_0} \sum_{\substack{|\mathbf{i}|=m-1 \\ i=i_0}} \sum_{|\mathbf{j}|=n} b_{\mathbf{j}} B_{\mathbf{j}}^n(\mathbf{i}/m) = \delta_{u,i_0}$$

after rearranging it:

$$\sum_{|\mathbf{j}|=n} b_{\mathbf{j}} \left(\frac{1}{m-i_0} \sum_{\substack{|\mathbf{i}|=m-1 \\ i=i_0}} B_{\mathbf{j}}^n(\mathbf{i}/m) \right) = \delta_{u,i_0}$$

similarly, for the trajectories where $v=j_0/(m-1)$ for some j_0 in $\{0,1,...,m-1\}$ and $w=k_0/(m-1)$ for some k_0 in $\{0,1,...,m-1\}$, we obtain:

$$\sum_{|\mathbf{j}|=n} b_{\mathbf{j}} \left(\frac{1}{m-j_0} \sum_{\substack{|\mathbf{i}|=m-1 \\ j=j_0}} B_{\mathbf{j}}^n(\mathbf{i}/m) \right) = \delta_{v,j_0} \text{ and}$$

$$\sum_{|\mathbf{j}|=n} b_{\mathbf{j}} \left(\frac{1}{m-k_0} \sum_{\substack{|\mathbf{i}|=m-1 \\ k=k_0}} B_{\mathbf{j}}^n(\mathbf{i}/m) \right) = \delta_{w,k_0}$$

These are the $3m$ rows of the overdetermined system $AX=b$, where X contains the unknowns $b_{\mathbf{i}}$. The least squares problem is set by the normal equations: $A^TAX=A^T b$.

The symmetric coefficient matrix of this linear system of equations possesses $(n+1)(n+2)/2$ rows and columns. Typically, when $n \leq \frac{\sqrt{24m+1}-3}{2}$, the least squares problem will return a unique solution.

Due to equation (2), and the fact that the Bernstein polynomials are non-negative, provided that the arguments are also non-negative, the error propagation is minimized when compared to other bases, such as the monomial or Lagrangean ones. Another advantage of this approach is the coordinate system: the gamma ray trajectories are specified by a constant (*w=k*, for instance) calling for less arithmetic operations than the affine equation *y=ax+b*. The homogeneity of the mini-triangles makes it easier to specify the intersections between the trajectories (*u=i/(m-1)*, *v=j/(m-1)* and *w=k/(m-1)*) benefiting in its turn each estimated trajectory mean density value. Yet another important characteristic is its symmetry in the domain. There is no privileged orientation, and the error is expected to be homogeneously distributed along the triangle area.

5. RESULTS

To implement the reconstruction technique, a computational algorithm by means of which catalyst concentration in static and flow experiments are presented, was written.

Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.

In the crown experiment a error of 13 % at the centre and of 5 % for the extremites were fond. In nucleus a 10 % error on the extremites and of 1% at center were evaluated .

6. CONCLUSIONS

To improve the least square approximation it is suggested to take the integral of density function along each trajectory, divided by the length of the trajectory inside the riser, instead of simply averaging the function values at the intersections between the trajectory with other trajectories. Another improvement may be to devise a method to estimate the function values in the trajectories, and then to use an interpolation method, such as a C^1 piecewise cubic Bézier triangles (Kong, 2004).

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