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The following report (which has s been submitted as a topical report) from Washington University for the period January -March 1996 contains the following chapters:

Abstract

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Slurry Bubble Column Hydrodynamics

Tracer Studies of the LaPorte AFDU Reactor During Methanol Synthesis

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Abstract

The radioactive tracer studies, using impulse injections of Mn^{56} 50 μm particles in the slurry phase and Ar^{41} in the gas, conducted at three process rates during the methanol synthesis runs : at the AFDU slurry bubble column reactor at LaPorte, Texas, were interpreted based on the axial dispersion model (ADM). Both liquid and gas axial dispersion coefficients showed an increase with superficial gas velocity. Responses of detectors located at various column heights point to the inadequacy of the ADM to interpret properly the gas and liquid flow pattern and mixing in the column. An alternative model is proposed.

Executive Summary

Radioactive tracer experiments were executed at the AFDU slurry bubble column reactor at LaPorte, Texas, at three different process rates (superficial gas velocities ranging from 14 to 36 cm/s, reactor temperature of 250 °C, reactor pressures of 3.6 MPa and 5.2 MPa, and a catalyst loading of approximately 45 wt %) to determine the flow pattern and backmixing of the liquid and gas during methanol synthesis. Impulse injections of radioactive Mn^{56} particles were used at different positions along the column to monitor the mixing of the liquid (batch) phase. Impulse injection of Ar^{41} were made at the syngas inlet to map the flow pattern of the gas. Responses to these tracer impulse injections were monitored at various column heights with four scintillation detectors located at 90° angles at each height. The axial dispersion model (ADM) was employed to interpret the tracer response data, since this model was used in the past for analysis of reactor performance.

The objectives of the study were : (a) to obtain flow pattern and backmixing information and backmixing data from actual pilot plant high pressure, high temperature reactors under operating conditions, (b) to examine the dependence of the obtained parameters on gas velocity and operating pressure, (c) to assess the ability of the available correlations to predict the measured axial dispersion coefficients and (d) to assess the suitability of the axial dispersion model for describing backmixing in reactors of this type and to determine the suitability of parameters used in reactor performance equations.

The findings, described and discussed in this report, can be summarized as follows:

1. The axial dispersion model (ADM) results in good fits with experimental tracer response data for the liquid phase. However, there is a wide scatter in the fitted axial dispersion coefficient, which points to the inadequacy of the model. An alternative phenomenological model is suggested.
2. The ADM is only fitted to the detector responses that showed no overshoots. The so-determined axial liquid dispersion coefficients typically exhibit a standard deviation of up to $\pm 35\%$.
3. Fitting of the ADM for the gas tracer response at detectors far removed from the inlet requires a good knowledge of both the gas holdup profile in the column and of the local gas velocity. Hence, tracer response is intimately tied to reactor performance which dictates the level of reduction in syngas flow rate depending on the level of conversion.

4. Questions can be raised about the accuracy of the measured holdup profile along the column height since differential pressure measurements are in considerable disagreement with nuclear density gauge measurements, and there is no independent evidence for the uniformity of slurry concentration along column height.
5. Fitting of the ADM to the gas phase response curves measured at a distance from the inlet, based on argon solubility calculated from thermodynamics, measured gas holdup profiles and assumed linear decay in gas velocity, yields the gas axial dispersion coefficients with a standard deviation typically within $\pm 22\%$. The overall volumetric mass transfer coefficient, which is the other parameter from such a curve fitting, exhibits a standard deviation of up to $\pm 100\%$.
6. The average value of the estimated parameters is shown in the table below:

Run	P	$\bar{\epsilon}_G$	U_{Gin}	X_{CO}	$D_L \pm \sigma_L$	$D_G \pm \sigma_G$	$K_L a \pm \sigma_k$
No.	MPa		cm/s		cm^2/s	cm^2/s	1/s
14.6	5.2	0.39	2.25	0.16	4355 ± 1532	5649 ± 1022	0.37 ± 0.37
14.7	5.2	0.33	1.14	0.18	2713 ± 678	2908 ± 723	0.38 ± 0.06
14.8	3.6	0.37	3.36	0.33	5143 ± 516	6621 ± 1437	0.35 ± 0.12

Both liquid and gas axial dispersion coefficients increase with increased gas velocity at constant pressure. The volumetric mass transfer coefficient seems unaffected by gas velocity. The effect of pressure is not clear since in run 14.8 while the pressure was reduced the superficial gas velocity was increased.

The available correlations for axial dispersion coefficients (with data base of atmospheric pressure operation) underpredict the observed liquid axial dispersion coefficient within 150% but overpredict the gas dispersion coefficient by 100% to 360%.

Additional work is needed in characterizing the reactors at operating conditions and in developing a suitable data base.

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1 Introduction

This investigation involves the study of the mixing characteristics in a slurry bubble column reactor during liquid phase methanol (LPMEOH) synthesis runs. The experiments were conducted in the AFDU Oxygenates High Pressure Reactor at LaPorte, Texas. Powdered methanol catalyst (~ 45 wt % loading) suspended in inert hydrocarbon oil forms the batch slurry phase. Synthesis gas is bubbled through a sparger placed at the bottom of the reactor. The gas disengages from the oil in the freeboard section of the reactor, and the unreacted feed gas is recycled back to the reactor.

The principal reaction for methanol synthesis is



At the process conditions used the methanol formed is in the vapor phase. The feed gas to the reactor is synthesis gas which is a mixture typically consisting of CO (30 %), H_2 (60 %), CO_2 (5 %) and inerts (N_2). The composition of the feed gas, given above in mole %, may be varied by changing the feed ratio, depending upon process requirements. The presence of CO_2 is usually required, as it serves to start the reaction. A side reaction known to occur is the water gas shift reaction:



Based on the above reaction stoichiometry (Equations 1 and 2), there is a reduction in the volume of the gas. The actual reduction depends on the feed rate, composition, and conversion. For the tracer runs studied, feeds with varying composition were used. The experimental conditions along with the feed compositions, observed conversions and changes in gas volumetric flowrate are reported in Table 1. The conversion of CO for the three runs studied ranges from 16% to 33%. An excess of CO results in lower conversion (runs 14.6 and 14.7 compared to run 14.8). Although CO conversion varies for the three cases, due to a corresponding change in feed composition, the effective overall change in the gas flowrate is about the same for all the runs, around -18 %.

Table 1: Experimental Conditions (Temp : 250° C)

Run No.	P MPa	Gas Holdup	U_G cm/s	Feed Composition mol %			Conv of CO to MeOH	Inlet Vol. Flowrate SCFH	Change in Flowrate %
				H_2	CO	CO ₂			
14.6	5.2	0.39	25	35.4	50.8	12.7	15.9	143121	-17.1
14.7	5.2	0.33	14	35.0	50.9	12.7	17.5	81151	-19.2
14.8	3.6	0.37	36	60.2	24.0	10.3	33.0	141690	-17.7

1.1 Gas Holdup Measurements

Holdup measurements within the reactor were made using two techniques. These are: 1. Differential Pressure measurements (DP) and 2. Nuclear Density Gauge (NDG) measurements.

DP measurements rely on the assumption that liquid (slurry) velocities and shear stresses near the wall are small in comparison with the hydrostatic head. Thereby

$$\rho g = \frac{\Delta P}{\Delta z} \quad (3)$$

where

$$\rho = \rho_L \epsilon_L + \rho_G \epsilon_G \quad (4)$$

The subscripts 'L' and 'G' refer to the slurry and gas phase, respectively. The density of the gas phase is very small when compared with that of the slurry, and therefore the second term in Equation 4 can be ignored. The slurry density in the column is assumed to be uniform (ρ_L) and is calculated using the information on solids holdup ((catalyst weight/density)/dispersion volume) (Shollenberger, 1995).

Nuclear Densitometry (NNDG) is a noninvasive method in which a source emitting a narrow beam of radiation through the column is used with a detector opposite it to scan across the column cross section. This yields a series of chordal measurements. A single line averaged holdup is usually obtained across the centerline, (i.e. diameter) of the column. However, this can result in a higher holdup value than the actual cross-sectional average holdup. A distribution of the void fraction across a given cross-section can be obtained in a series of such scans at different angular orientations (tomography), which on averaging results in the true cross-sectional mean. The difference between the holdup estimates from these two γ ray techniques (i.e., tomography and chordal averaging) is illustrated in Figure A.1.1. of Appendix I. The reasons for the discrepancy have been discussed elsewhere (Kumar et al., 1996).

It was found that the nature of the averaging method in the NDG measurements leads to higher values of gas holdup than the actual cross-sectional average value, especially for cases of steep void fraction profiles in the column. However, when the void fraction profile is more uniform, the discrepancy between DP and NDG determined holdup values is reduced. Due to this factor, for present considerations, the DP measurements will be initially considered for more accurate holdup estimates.

Axial holdup profiles at various process conditions are shown for both measurements in Appendix I (Figure A.1.3.). It is seen that there is an initial decrease in holdup with axial position, z , up to a height of about 10 column diameters, after which the holdup remains quite uniform up to a height of about 15 column diameters. From this level upward there is a steady increase in gas holdup. Since the reaction results in the decrease of gas volume, it is surmised that this increase in holdup towards the free surface of the liquid is the result of foaming in the system. Such a trend in the axial gas holdup profiles is reported for all process conditions (Figure A.1.3.a).

Unfortunately, even this result of increasing gas holdup with column height in the upper sections of the column cannot be unconditionally accepted at face value. If a catalyst concentration gradient develops along this tall column, then the slurry concentration at the top of the column is smaller than the one at the bottom (or the average), i.e., $\rho_{L,T} < \rho_L$. The pressure drop measurement by the DP method in the top region of the column actually yields

$$\left(\frac{\Delta P}{\Delta z}\right)_{top} = \rho_{L,T} \epsilon_{L,T} g \quad (5)$$

However, if we use the average slurry density (ρ_L) to estimate the slurry holdup, ϵ_L from this measured value, it can be readily seen from Equation 6 that the slurry holdup is underestimated since $\rho_{L,T}/\rho_L < 1$.

$$\epsilon_L = \frac{1}{\rho_L g} \left(\frac{\Delta P}{\Delta z}\right)_{top} = \epsilon_{L,T} \frac{\rho_{L,T}}{\rho_L} < \epsilon_{L,T} \quad (6)$$

Therefore the gas holdup is overestimated, and the rise of gas holdup with height in the upper part of the column may be the artifact of the DP measurement interpretation. It should be noted that if NDG is used, and the attenuation of the slurry varies with catalyst concentration, then a less dense slurry at the top of the column, if the attenuation calculation is based on a more concentrated slurry, will yield an overestimate of gas holdup also.

This indicates that both the DP and NDG techniques can yield, as an artifact of the assumption of uniform catalyst concentration, an increased gas holdup along the

column length if pronounced axial catalyst concentration profiles are present.

2 Objectives

The objective of this work is to study the mixing characteristics of the gas and liquid phase in the slurry bubble column reactor using radioactive tracer experiments. Interpretation of the tracer experiments at this stage is based on the one-dimensional axial dispersion model (ADM). The ADM is chosen because it is almost always used by others, and since the previous runs at LaPorte have utilized this approach as well. This choice is, in addition, dictated by the fact that ADM has also been used in reactor design and performance calculations. Thus, examination of the tracer studies can provide some insight regarding the appropriateness of the backmixing parameters used in reactor performance calculations.

However, our long-term goal is to assess the suitability of the present model (ADM) to describe mixing in the gas and liquid phase in slurry bubble column reactors and suggest alternatives if needed.

3 Tracer Experiments

A schematic of the AFDUU slurry bubble column reactor is shown in Figure 1. It has an internal diameter of 0.46 m and a height of 15.24 m, with the liquid - gas - solid dispersion level maintained at 13.25 m (L/D ratio of 28.8) during the runs discussed here. The vapor phase and liquid phase tracer experiments were conducted separately. Radioactive Ar-41, used to study the residence time distribution of the vapor phase, was injected as a pulse at the inlet of the reactor. Radioactive Manganese-56 (50 μm) particles mixed in oil were used for liquid (slurry) phase tracing. Four pulse injections were made at a given process rate: (1) lower nozzle N2 - 4.5" (11.4 cm) from wall, (2) nozzle N2 - at wall, (3) upper nozzle N1 - 4.5" (11.4 cm) from wall, and (4) nozzle N1 - at wall. The axial levels of these injection points are shown in Figure 1. The injections made at 4.5" (11.4 cm) from the wall will be referred to as "center injections," as they are made into the core part of the column where the liquid is known to move upward by convection (Devanathan et al., 1990).

Radiation measurements from the vapor and liquid tracers were made using several 2" by 2" NaI scintillation detectors positioned outside the column, at various axial levels, as shown in Figure 1.1. Sets of four detectors were placed at 90 degree angles at seven heights. In addition, detectors were placed at the inlet and outlet of the

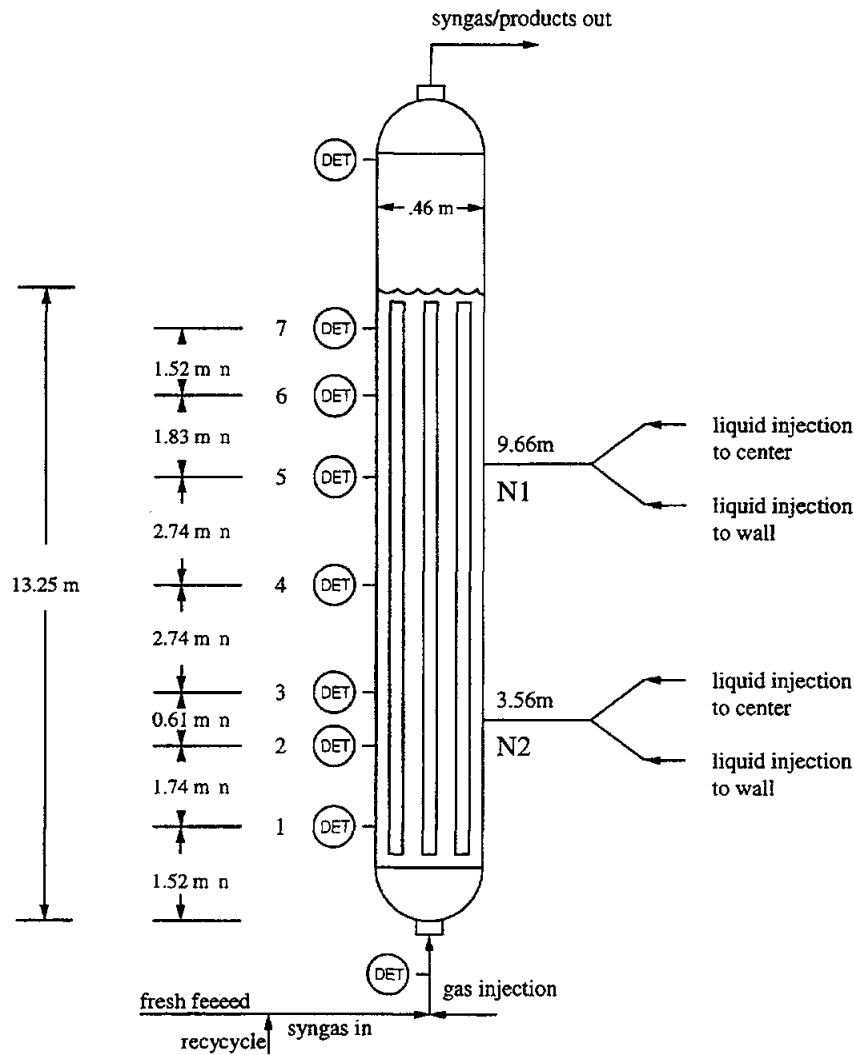


Figure 1: Schematic of Reactor for Tracer Experiments

reactor. During the liquid tracer study, the inlet detector was placed close to the liquid injection point to monitor the shape of the injected pulse.

The radiation (counts/time) measurements made at the detectors were used to obtain the residence time distribution of the radioactive tracer. To analyze of the tracer data with the one dimensional axial dispersion model (ADM) for both the vapor and liquid phase, the responses from the four detectors at each level were averaged to obtain a cross-sectional averaged response at the corresponding level. This was done since the model is one dimensional and does not have the capability of resolving radial and azimuthal variations.

The total radiation (counts/time) recorded at the detector is an integral of the

contribution of the entire mass (or volume) of tracer, which can be considered to comprise individual point sources within the field of view of the detector. For the tracer experiments considered in this study, the detectors were shielded on their sides. Therefore only the front circular surface of the detectors could see the radiation. With this configuration, the spatial range from which a detector receives most of its signal was assessed. The equations used for this calculation (Tsoufanidis, 1983) are shown in Appendix II. For a uniform distribution of radioactive tracer in the column, it is concluded that more than 90 percent of the intensity recorded at a detector, shielded on its sides, comes from a small volume (less than 1 percent of the entire reactor volume) closest to the face of the detector. This is shown in Figure A.2.1, where the exponential decay in intensity with distance can be seen.

For gases and liquid, which are no longer point sources, the specific activity, defined as the number of disintegrations/time/volume of the nuclei of the radioisotope, depends on the mass or volume of the radioactive source (Tsoufanidis, 1983), and thereby on the local concentration of tracer.

Since the ADM, when applied to the liquid and gas phase, can only consider uniform tracer concentration and phase holdups, the intensity (counts/time) recorded at the detector can be assumed to be directly proportional to the concentration of tracer at a given axial location of the detector. The liquid Mn-56 tracer with a half life of 2.58 hr emits γ radiation at 0.85 MeV. This tracer remains only in the liquid (slurry) phase. Hence, the average detector response is directly proportional to the tracer concentration in the liquid phase. However, the gas tracer Ar-41 of half life 1.29 hr, emitting γ rays at 1.29 MeV, is soluble in the liquid. Therefore, for the gas experiments, the total tracer concentration at a given time and axial position, which is assumed to be proportional to the average detector response, is taken to be

$$C_{t_i}(t, z) = \epsilon_L C_L(t, z) + \epsilon_G C_G(t, z) \quad (7)$$

4 Modeling

The one dimensional axial dispersion model (ADM) was used to model mixing in the liquid and vapor phase. It was chosen for reasons stated earlier. This model essentially assumes the fluid to be in plug flow with axial dispersion superimposed on it.

4.1 Liquid Phase Tracer

For the case of a batch liquid (slurry) with non-volatile tracer, the one dimensional dispersion model can be written as:

$$\frac{\partial C_L}{\partial t} = D_L \frac{\partial^2 C_L}{\partial z^2} \quad (8)$$

Initial and boundary conditions are:

$$t = 0; \quad C_L = \delta(t)\delta(z - z_i) \quad (9)$$

$$z = 0; \quad z = L; \quad \frac{\partial C_L}{\partial z} = 0 \quad (10)$$

where z_i and L denote the level of injection and liquid dispersion height, respectively. The model has a single parameter, the liquid phase dispersion coefficient, D_L . It can be solved analytically, and the response at a given detector level, z , to an impulse input at z_i is:

$$C_L(t, z) = 1 + 2 \sum_{n=1}^{\infty} \cos\left(\frac{n\pi}{L} z_i\right) \cos\left(\frac{n\pi}{L} z\right) \exp(-D_L n^2 \pi^2 t) \quad (11)$$

The average response of the four detectors at each level was considered. Both experimental data and model predicted responses were normalized with respect to their maximum value before the fitting procedure was performed. Hence the best value of the parameter, D_L , was obtained by minimizing the sum of the square of the errors, given by

$$\min_{D_L} \sum_{i=1}^{N_p} \left(\frac{R(t_i, z)}{R_{max}} - \frac{C_L(t_i, z)}{C_{Lmax}} \right)^2 \quad (12)$$

where $R(t_i, z)$ is the averaged response of the four detectors at z and time t_i , and R_{max} is the maximum value of the detector response at z . $C_L(t_i, z)$ is the model predicted liquid tracer concentration at level z and time t_i , and C_{Lmax} is the maximum liquid concentration. The number of data (sampling) points, N_p , is about 1000.

Time domain fitting of the model prediction with the average detector response at every level is performed, for each injection, to estimate the model parameter D_L . It is noted here that the tracer injections are point injections, not cross sectional injections, and therefore do not truly satisfy Equation 9 (uniform injection in a cross section), since a finite time is required for the tracer to spread radially.

4.2 Gas Phase Tracer

Since the radioactive argon tracer is soluble in the liquid phase, the one dimensional axial dispersion model with interfacial mass transfer resistance, is considered for modeling the argon tracer distribution in the reactor. The mass balance equations for the tracer in the vapor and liquid phase are as follows :

$$\frac{\partial C_G}{\partial t} = D_G \frac{\partial^2 C_G}{\partial z^2} - \frac{U_G}{\epsilon_G} \frac{\partial C_G}{\partial z} - K_L a \left(\frac{C_G}{H} - C_L \right) \quad (13)$$

$$\frac{\partial C_L}{\partial t} = D_L \frac{\partial^2 C_L}{\partial z^2} + \frac{\epsilon_G}{\epsilon_L} K_L a \left(\frac{C_G}{H} - C_L \right) \quad (14)$$

The initial and boundary conditions are :

$$t = 0, \quad C_L = 0; \quad C_G = 0 \quad (15a)$$

$$z = 0, \quad \frac{\partial C_L}{\partial z} = 0; \quad \frac{U_G}{\epsilon_G} C_G = D_G \frac{\partial C_G}{\partial z} + \frac{U_G}{\epsilon_G} \delta_i(t) \quad (15b)$$

$$z = L, \quad \frac{\partial C_L}{\partial z} = 0; \quad \frac{\partial C_G}{\partial z} = 0 \quad (15c)$$

D_G is the gas phase axial dispersion coefficient, D_L is the liquid axial dispersion coefficient, H is Henry's law constant for the gas tracer, $K_L a$ is the overall volumetric gas-liquid mass transfer coefficient based on the volume of gas in the reactor, U_G is the inlet superficial gas velocity, and ϵ_G and ϵ_L are the gas and liquid (slurry) holdups, respectively. The function $\delta_i(t)$ describes the pulse of tracer injected and has an area under the curve of unity. For a perfect impulse injection $\delta_i(t) = \delta(t)$ where $\delta(t)$ is the Dirac delta function.

It should be noted that Henry's constant in this study is defined as

$$H = \left(\frac{C_G}{C_L} \right)_{eq} \quad (16)$$

and represents the ratio of the argon molar concentration in the gas and liquid phases when the two phases are in equilibrium. Thus H , as defined in this report, is dimensionless. This can be readily related to the more customary expression

$$H' = \left(\frac{p_G}{C_L} \right)_{eq} \quad (17)$$

by using an equation of state to relate the partial pressure of the tracer gas, p_G , to its gas phase concentration C_G . If C_L is measured in (mol/m^3) and p_G in atmospheres, the units for H' are ($\text{atm nm}^3 / \text{mol}$).

The overall liquid volumetric mass transfer coefficient is defined by

$$\frac{1}{K_L a} = \frac{1}{k_L a} + \frac{1}{H' k_G a} \quad (18)$$

where k_L (m/s) is the liquid side mass transfer coefficient, while k_G ($\text{mol}/\text{m}^2 \text{ s atm}$) is the gas side mass transfer coefficient. For slightly soluble gases the second term in Equation 18 can be ignored, and liquid side resistance is dominant. The interfacial area, a , as used in the present model, is the gas-liquid area per unit volume of the gas.

We have constrained ourselves here to examining the ADM with constant holdup and constant gas superficial velocities as this has been traditionally used for assessment of reactor performance. We wanted to determine whether such a model can provide a consistent set of reactor parameters when fitted to observed tracer responses.

The model was solved numerically using NAG subroutines for solving the system of partial differential equations. Basically, if one assumes that D_L is known from liquid phase experiments, there are five model parameters, D_G , ϵ_G , H , $K_L a$ and U_G . Three additional parameters are available from independent measurements or calculation: ϵ_G is known from DFP (and NDG) measurements, and H can be evaluated from thermodynamics. The superficial velocity at the inlet and outlet is known. In this report, six different parameter estimation trials were conducted in order to estimate the parameters of the model. The details and reasons for this will be discussed in the next section.

Impulse response measurements were performed for the Argon-41 tracer injected into the vapor phase. The tracer was injected as a pulse into the inlet gas line upstream of the sparger. A detector was placed approximately 10 cm (which is close) from the injection point, upstream of the reactor, to monitor the nature of the input pulse. Thus, the detected pulse did not necessarily describe the true shape of the input at the inlet of the reactor since it was detected prior to the inlet. However, due to lack of any better information, this was used to simulate the input pulse to the reactor for the purpose of modeling.

The observed input tracer pulse for the model can be represented well by the following Gaussian function, with adjustable variance to match it to the response of

the inlet detector:

$$\delta_i(t) = \frac{N_c}{\sqrt{2\pi D_i t}} \exp\left(-\frac{(l_i - U_G t)^2}{2D_i t}\right) \quad (19)$$

where D_i and l_i are the parameters used to match the simulated pulse with the measured response of the inlet detector, and N_c is the normalization constant. This model provides good fits to the inlet detector response, i.e., the input pulse to the reactor. (see Figure 2 for an example - run 14.7-1. Fits for other process rates are in Appendix VI.) Since the model responses and parameters were found to be sensitive to the input function used, care should be taken when simulating the input pulse for a given experiment.

As in the case of the liquid tracer data, the detector responses and the model predictions were normalized with respect to the maximum height of the response curve before the model response was fitted to data.

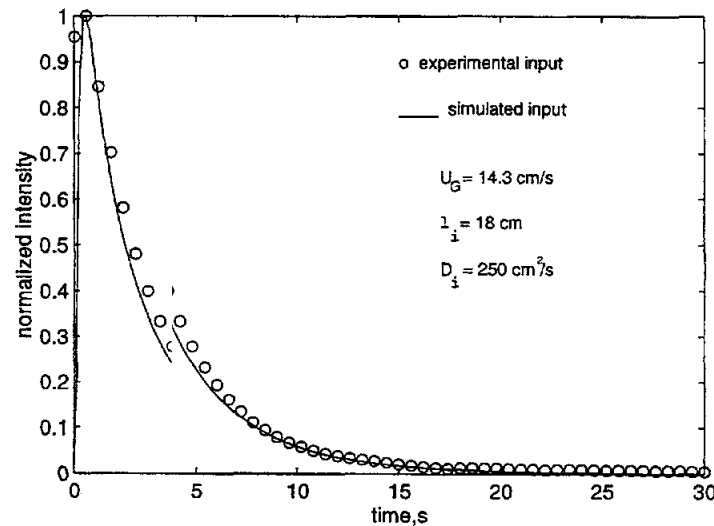


Figure 2: Simulation of Input Pulse to the Reactor

5 Parameter Estimation by Fitting the Model to Data

5.1 Liquid Tracer

Four liquid tracer injections were made at two axial levels for each process rate. For an injection at a given level, responses of the detectors closest (two levels on

either side) to the point of injection showed overshoots and lack of symmetry (see Figure 3, where 'CEN' refers to the center injection). Presently, the model is used to fit only the data of the detectors located at the three levels farthest away from the point of injection that do not exhibit overshoots. The results of the fitting (D_L), for all cases, are tabulated in Table 2. Typical fits to the experimental data are shown in Figure 4 for run 14.7 - N2 center injection. The actual starting time for each experiment (injection) was different from (later than) the zero time of measurement for the detector counts. This was reflected in the offset in the time axis of the tracer response curves shown. The actual injection times were known from the response of the detectors at the point of injection. In the model the time is counted from the injection time. All figures, however, show a brief history prior to injection. The graphs for all the remaining experiments are shown in Appendix III.

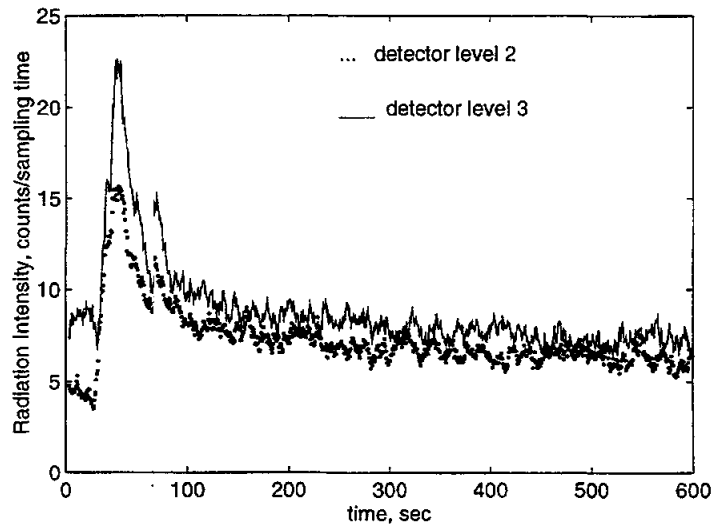


Figure 3: Impulse Response Measurements for N2 CEN Injection

5.1.1 Discussion

The fits of the model to experimental responses are, in general, good. The following observations can be made, regarding the values of D_L obtained:

- Level of injection - NN1 and N2 :

The values of D_L obtained from the top injection, i.e., injection nozzle N1, are different from those obtained by fitting the tracer data from the bottom injection, nozzle N2. This suggests the nonsymmetric axial distribution of the tracer about its point of injection, contrary to the nature of the ADM.

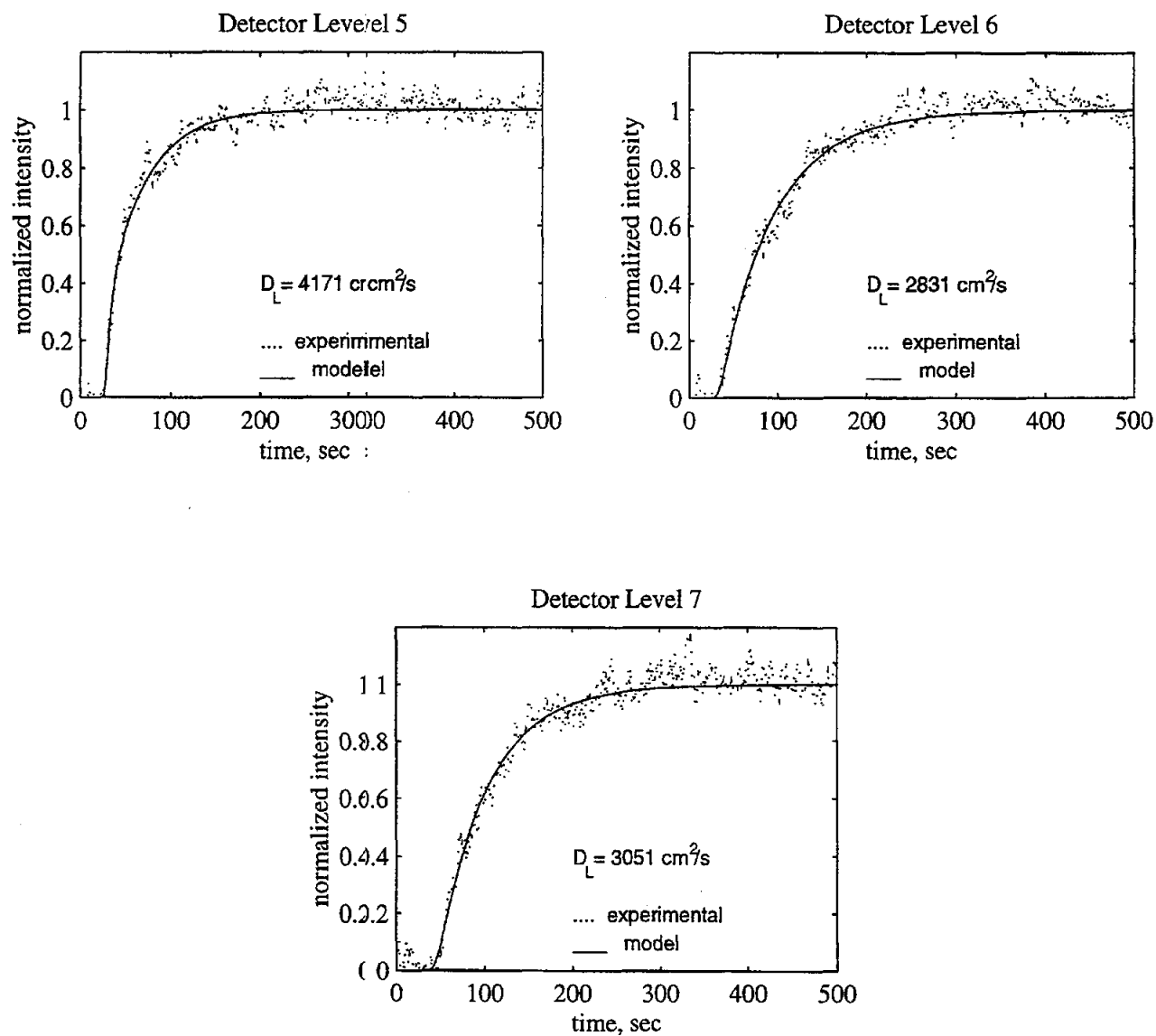


Figure 4: Liquid Impulse Response Measurements for Run 14.7, N2-CEN Injection, Injection Time: 7.8 s