II. DESCRIPTION OF EXPERIMENT

A. Gas Tracer

Mixing studies were performed on the LPMEOH reactor located in LaPorte, Texas. The reactor is a stainless steel column with an internal diameter of 22.5 inches and a possible liquid height of 25 feet. The slurry phase is stationary and contains an inert hydrocarbon oil and a powdered methanol synthesis catalyst. Synthesis gas is bubbled through the slurry via a gas sparger. Oil becomes disengaged from the gas in a space above the liquid and the gas exits from the reactor through the top of the column. A detailed description of the LPMEOH PDU has been presented in several previous DOE program reports [23,24] and will not be repeated here.

The gas phase residence time distribution was measured by ICI TracerCo of Houston, Texas. For each experimental condition, the gas holdup, liquid level, temperature, pressure and gas flow rate were measured prior to injection of the radioactive tracer. Argon (Ar-41) was irradiated at Texas A&M University and transported to LaPorte on the day of the test. Radioactive Ar-41 (chosen for its half-life of 1.8 hours) was transferred to a gas sampling bomb which was connected to the gas feed line through a sampling valve near the reactor bottom. At the start of the experiment, the contents of the bomb were injected into the gas feed line using a purge of high pressure nitrogen. The radioactive gas was vented to the atmosphere in accordance with Texas state law.

Twenty detectors were located around the reactor to measure the response for 10 minutes after the injection. The most important detectors were on the inlet, outlet and directly above the slurry level. This allowed measurement of the residence time distribution (RTD) for the entire reactor and also the RTD of the gas in the slurry portion of the reactor. Mixing of the gas in the liquid portion of the reactor is of most interest because that is where the catalyst is located and the reaction occurs. The response was recorded digitally at a rate of 4000 points per 10 minute period per detector. At least three injections were made at each condition to check consistency of results. After the injections were made for a given condition, the gas holdup, liquid level, temperature, pressure and gas flow rate were measured.

Over a two day period, six conditions were studied for the gas tracer as shown by the first six entries of Table II.A-1. Three gas velocities were studied at full slurry height and three gas velocities were studied at half slurry height. This information was used to determine mixing effects as a function of L/D and of gas velocity.

Table II.A-1

Nominal Conditions for Tracer Studies

Tracer	Case	Inlet Gas Velocity (ft/sec)	L/D
Ar-41	1	.25	10
Ar-41	2	.5	10
Ar-41	3	.6	10
Ar-41	4	.5	6.5
Ar-41	5	.25	6.5
Ar-41	6	.18	6.5
Mn_2O_3	7	.5	11

T = 250°C
P = 750 psig

B. Liquid Tracer

The slurry mixing test was also performed by ICI TracerCo. Manganese oxide (Alpha Chemicals, Mn_2O_3, 98% pure, -325 mesh) was supplied to Texas A&M for irradiation. Mn_2O_3 was selected as the tracer since it would not interfere with the methanol reaction. A particle size less than 44 microns was chosen to match the size of the catalyst particles. The degree to which suspended particles follow turbulent liquid mixing a bubble column is discussed in a recent paper by Matsumoto et al.[9] the basis of mixing length theory. For the present case, the particle Reynolds number for a 44 micron particle, based on the fluctuation velocity, is about 6. This number is defined by

$$Re_p = \frac{d_p\,(gD\varepsilon_g)^{1/2}}{\nu_L\sqrt{2}} \qquad (1)$$

(written here for the case of zero bulk liquid velocity). The ratio of the mixing time associated with a turbulent fluid eddy to the response time of a suspended particle is characterized by the group β defined below. A large value of β would indicate that the response time of the particle is small compared with the mixing time of the fluid.

$$\beta = \frac{\varnothing\; L\,\rho_L}{(1-\varepsilon_g)\,\nu_L\,\rho_p\,Re_p^2} \qquad (2)$$

$\mathcal{D}_L$ is the liquid dispersion coefficient based on total volume. For our case, β is about 400 for a 44 micron particle, and it is even larger for smaller particles. In this range, it is clear that the particle closely follows the fluid.

A pretest was performed by Texas A&M to verify the half-life of 2.5 hours. Unlike the gas tracer, the solid powder would remain in the reactor, so a short half-life was important. Since the tracer remained in the reactor, only one injection per day was permitted by Texas state law.

Approximately 0.1 gm of radioactive Mn_2O_3 was loaded into a gas sample bomb and mixed with reactor oil. The gas bomb was connected to a 2" reactor inlet pipe at the bottom of the reactor. Although DME Solid Tracer Studies done in the past have injected the powder at the top of the reactor, the current reactor configuration would not allow this. For future tracer study tests, modifications to the reactor will be considered which would allow this method of testing. Experimental conditions were measured before and after the injections as was done in the gas tracer tests. The conditions for the liquid tracer test are listed in **Table II.A-1**. At the start of the experiment, the contents of the bomb were injected into the reactor using a purge of high pressure nitrogen. Twenty detectors were located around the reactor to measure the response for 10 minutes after the injection.

C. Run Summary

The cases in **Table II.A-1** were performed during February and March of 1989. On February 24, 1989 cases 1, 2, and 3 were conducted at full slurry level. To maintain a constant concentration of solids, the slurry level had to be varied slightly with gas flow rate. Three injections were made for each case. Due to equipment problems during case 3, four injections were made. On February 28, 1989, cases 4, 5, and 6 were conducted at the lower slurry level. The liquid tracer, Case 7, was performed on March 30, 1989. Detectors 3, 5, 6, 7, 8, 9, 13, and 16 malfunctioned and the injection was repeated by blowing in the remaining Mn_2O_3 that lay in the injection line. The second injection did not contain enough radioactivity to provide a strong signal for the detectors. However, enough detectors had worked during the first injection to provide data at each reactor level. Additional Mn_2O_3 injections were not possible on March 30 due to a state-imposed limit on radiation dosage. Actual conditions for all these cases are listed in **Table II.C-1** and the position of the detectors for each case is reported in **Appendix A**.

D. Data Reduction

The data analyzed in this report consisted of 4000 points per detector taken over a ten minute time span for each injection. During each injection, there were 20 detectors mounted on the reactor at various elevations. Each point represents the relative amount of radioactive material that was present at that position in the reactor at that instant in time.

The original data were received in the form of 21 sets of numerical strings roughly 1.4 megabytes in length. Each continuous numerical string represented a single injection and consisted of 4000 points from each of the 20 detectors. This initial data set was reduced to a set of 20 files per injection with 4000 points per file.

Figure II.D-1 shows a detector file plotted on a real time scale. Notice that the radiation increases at long times. At the LaPorte PDU, a portion of the gas product is recycled and mixed with fresh syngas feed. The "tail" seen in the detector data is due to radiation returning in the recycle; it was omitted in the data analysis. Many of the response curves reached a baseline that was less than 10% of the maximum value before the recycle radiation was detected. Depending on the type of curve, omitting the tail could introduce a 10% error in the normalization factor if the mixing is characterized by a single CSTR, or as low as a 3% error if the mixing is plug flow. The plots of all the data resulted in a thick curve that was very difficult to integrate and fit to a model because of the "noise" and the number of data points involved.

Therefore, a method was required to normalize, and if possible, smooth each response curve. The three different types of curves that were analyzed were the impulse spike, the gas phase detector response curves, and the liquid phase detector response curves.

The first five minutes of data from the impulse spike was integrated using the trapezoidal rule and the baseline (background) radiation determined. The baseline radiation was subtracted from the data points and the data were normalized by dividing the value by the peak area.

The noise in the gas phase response curves was eliminated by using the Fourier transform. The Fourier transform converts the data from the time domain to the frequency domain. With the data in the frequency domain, the high frequency points (noise) were omitted and the inverse Fourier transform was performed. The resulting smooth curve overlays the original curve and has a low degree of noise (Figure II.D-2). With the data in this form, it was possible to subtract the baseline radiation from the data. The data were integrated using the trapezoidal rule and normalized using the following equation:

$$\text{norm}_i = (X_i - \text{Baseline})/(\text{Area} - \text{Baseline} * \text{No. data points})$$

Where norm_i is the normalized data, X_i is the transformed data, "Area" is the integrated area under the transformed curve and "Baseline" is the baseline radiation.

Similar techniques were used to reduce and normalize the liquid tracer data. However, since the tracer powder remained in the reactor, the radiation counts leveled out, not at zero as in the gas phase tests, but at a value which represents a well-mixed system. Therefore, the response curve was not normalized by the integrated area, but rather by the value termed well-mixed so that the curves leveled out at a value of 1.0.

Table C-1

Actual Conditions for Tracer Studies

Case	Injection Number	Slurry Level (tape)* at Injection	Gas Flow scfh	Inlet Superficial Gas Velocity ft/s	Ave. Temp F	Ave. Press psig	Fraction Gas Holdup	Catalyst Inventory Kg	wt % Cat (Oxide)
1	1	185	73040	.25	482	753	.296	362	36.3
	2	185							
	3	184							
2	4	206	146640	.50	483	753	.363	362	37.
	5	205							
	6	205							
3	7	210	175940	.60	484	753	.378	362	36.4
	8	207							
	9	207							
	10	205							
4	b1	135	145210	.50	481	753	.353	256	37.2
	b2	135							
	b3	134							
5	b4	117	72640	.25	482	753	.273	256	37.5
	b5	115							
	b6	115							
6	b7	108	54370	.18	487	753	.227	256	37.5
	b8	108							
	b9	108							
7	L1	216	146600	.50	482	753	.439	301	34.2
	L2	210							

* The zero of the tape measure is 32" above the bottom of the reactor.

Figure II.D.1 LAPORTE LPMEOH TRACER STUDY
TYPICAL RADIATION COUNTS vs TIME FOR REACTOR OUTLET

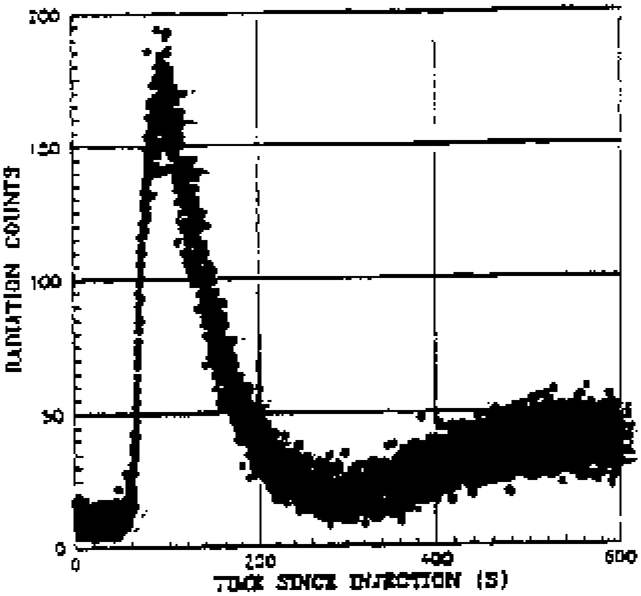

FIGURE II.D.2 : LAPORTE LPMEOH TRACER STUDY
NORM. RADIATION COUNTS vs TIME AT REACTOR OUTLET