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Project Summary

Coal Gasification/Gas Cleanup Test Facility: Volume IV. A Mathematical Model of the Packed Column Acid Gas Absorber

R. M. Kelly, R. W. Rousseau, and J. K. Ferrell

The report describes a mathematical model for adiabatic operation of a packed-column absorber designed to remove acid gases from coal gasification crude product gas. It also gives results of experiments with a small pilot-scale coal gasification/gas cleaning facility designed to test the model. The model predictions compared well with the actual absorber liquid temperature profile and outlet gas composition. The model is useful for the evaluation of the effect of changes in process variables on absorber column performance and (hence) for column design.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

As a part of a continuing research program on the environmental aspects of fuel conversion, the U.S. Environmental Protection Agency (EPA) has sponsored a research project on coal gasification at North Carolina State University. The facility used for this research is a small coal gasification/gas cleaning pilot plant. The overall objective of the project is to characterize the gaseous and condensed phase emissions from the gasification/gas cleaning process, and to determine how emission rates of various pollutants depend on adjustable process parameters.

The plant, described in detail in Volume I of this report series, consists of a fluidized bed reactor; a cyclone and venturi scrubber for particulates, condensables, and solubles removal; and absorption and stripping columns for acid gas removal and solvent regeneration. The plant has a nominal capacity of 50 lb*/hr of coal feed for steady state operation. A schematic diagram of the gasifier, the acid gas removal system (AGRS), and other major components is shown in Figure 1.

In an initial series of runs on the gasifier, a pretreated Western Kentucky No. 11 coal was gasified with steam and oxygen. The results of this work and a detailed listing of the project objectives are given in Volume II and were presented at the EPA Symposium on Environmental Aspects of Fuel Conversion Technology V, in St. Louis MO, September 1980.

Volume III gave a detailed discussion of a series of runs made with a New Mexico subbituminous coal. Part of Volume III summarized a mathematical model of the operation of the packed tower absorber of the acid gas removal system. This model may be used to estimate the height of packing required to remove a specified component from a gas stream or recover a specified component for a column with

(*) Readers more familiar with metric units may use the conversion factors at the end of this Summary.

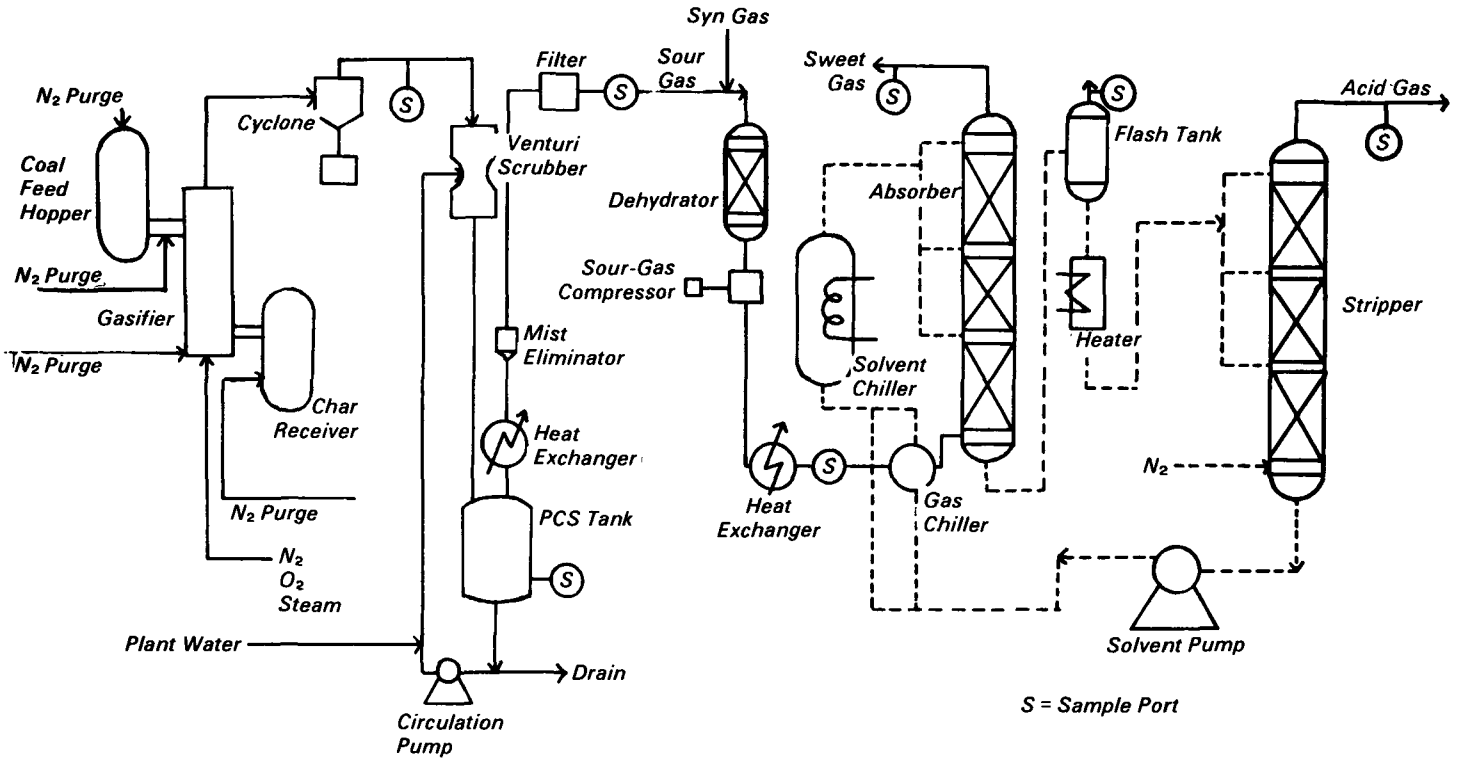


Figure 1. *Pilot plant facility.*

a fixed packing height. Furthermore, parametric studies involving a variety of process variables can be performed which may lead to an improved design and/or operation.

This report gives a detailed description of the absorber model and its development, and compares model-based predictions with experimental data from the pilot plant.

Packed Absorption Column Model

Several assumptions typically have been made in developing a physical model for non-isothermal absorption in a packed tower. Since axial dispersion is usually neglected, fluids are assumed to move through the column in plug flow. This is probably valid for small diameter columns with low to moderate liquid flow rates. Another assumption often made is that the column approaches adiabatic operation; most industrial columns have been shown to operate nearly adiabatically. Departures from adiabatic operation, due to either heat losses or planned heat removal, can be included in the model on a case-by-case basis.

Figure 2 is a schematic of a packed tower for which the following differential material and energy balances may be written:

$$\partial L_i/\partial z - \partial G_i/\partial z = \partial b_i/\partial t \quad (1)^*$$

$$\partial (LH_L)/\partial z - \partial (GH_G)/\partial z - H_{ex} = (bH_b)/\partial t \quad (2)$$

The resulting system of coupled, partial differential equations is very difficult to solve, and several approaches are possible. At steady state, Equations 1 and 2 become ordinary differential equations and, if adiabatic operation is assumed, the following equations may be written:

$$dL_i dz - dG_i/dz = 0 \quad (3)$$

$$d(LH_L)/dz - d(GH_G)dz = 0 \quad (4)$$

The steady state assumption leads to a two point boundary value problem with neither boundary condition completely specified. One solution involves an iterative calculation in which the conditions at one end of the column are assumed and an incremental calculation is initiated at the other end. When the calculation reaches the end of the column where the initial assumptions were made, calculated and assumed values are compared. If agreement between these values is unsatisfactory, an iterative procedure must be used to adjust assumed values. Algorithms have been suggested for both single-solute and multicomponent systems, although experimental verification of only the former has been reported.

Aside from the assumptions that the column is operating adiabatically and the axial dispersion is negligible, the single-solute calculational procedure is essentially rigorous. While more simplifying assumptions could be made (e.g., ignoring the resistance of one phase to mass and heat transfer), they could lead to serious error in the calculation. Certain situations may allow a less rigorous approach, but it is often difficult to determine this without first performing a more detailed calculation.

Description of Computer Programs

Two separate but similar computer programs were used in this study. SIMPAK was limited to the simple case of a single transferring solute in an inert, insoluble carrier gas and a volatile

(*)Nomenclature of equation elements is defined at the end of this Summary.

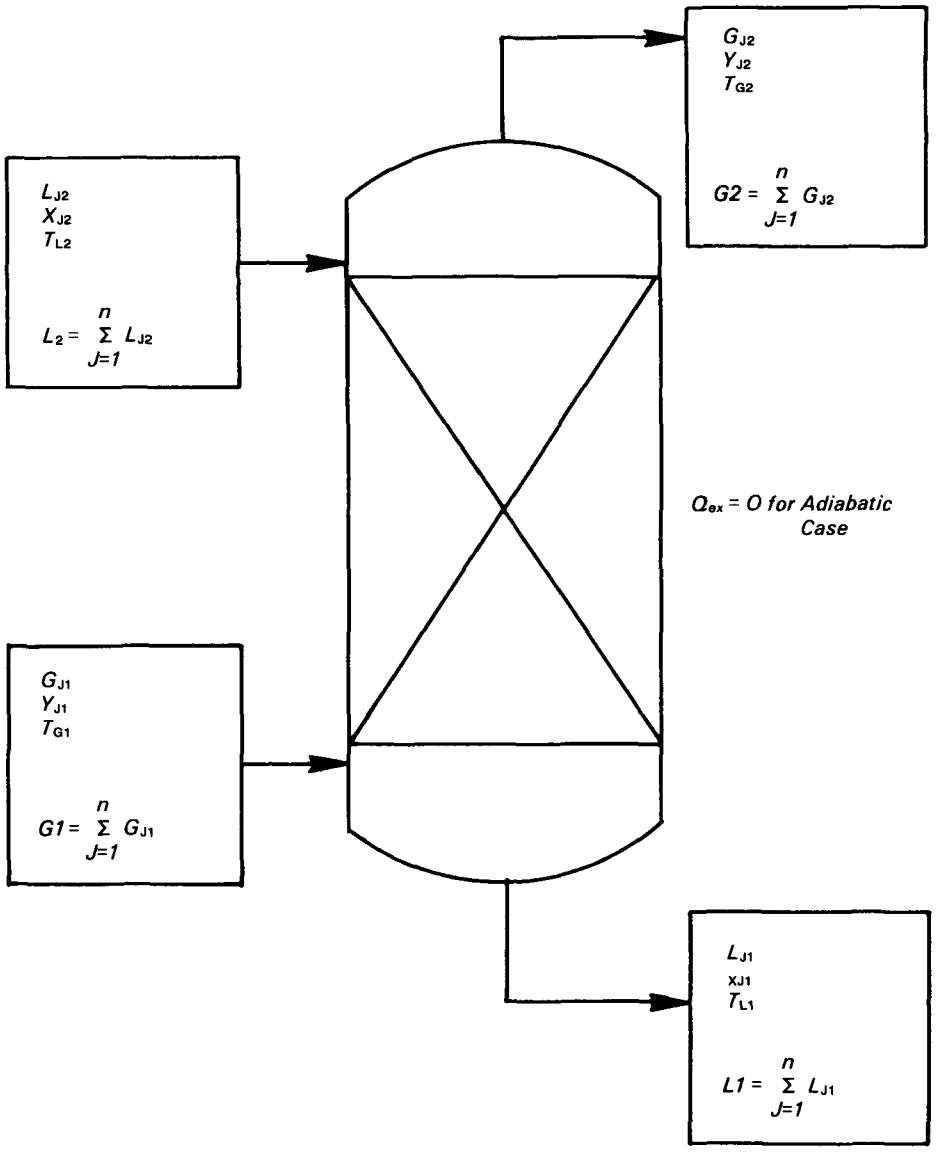


Figure 2. *Multicomponent gas absorption/stripping in an adiabatic packed column.*

solvent, while MCOMP relaxed these restrictions.

SIMPAK receives as input the condition and composition of the entering gas and liquid streams and the specified removal efficiency of the key component. The key component can be chosen from Table 1, a list of 13 compounds. Methanol, the liquid used in this study, must be designated as the solvent. The inert carrier gas can also be any compound in Table 1, except methanol. In general, the key component should be appreciably soluble in methanol, while the inert component should be sparingly soluble in methanol.

For multicomponent absorption, the computer program MCOMP has been developed. The program is a modification of the computer code used by H. M. Feintuch. Although any number of components can be handled, the 13 Table 1 components, designated as the most important in coal gasification applications, have been made a part of the model.

Because any of the 13 components can transfer in either direction, the problem becomes much more complicated than for single-solute absorption. Although the algorithm used in this case is similar to SIMPAK, including multicomponent mass transfer requires certain modifications.

After input of all necessary information (see Table 2), the calculation is initiated by assuming the outlet gas temperature and outlet gas flow rates for all components, except the key component, which has been specified. The outlet gas temperature is assumed initially to be equal to the inlet liquid temperature. After the flow rates of all exiting gas stream components are assumed, material and energy balances are solved to determine the conditions of the exiting liquid stream.

The calculation then begins at the bottom of the column. The total amount of key component to be removed in the column is divided by the number of packing segments to be used. The height required to achieve the removal of the key component in each segment is determined by calculating the temperature and concentration gradients.

Physical, Transport, and Thermodynamic Properties

The success of any modeling hinges on either the availability of accurate physical, transport, and thermodynamic property data or the ability to predict this information. Surprisingly, relatively little effort has been invested in expanding existing physical property data bases and improving predictive methods. This is especially true for mixture properties and at other than ambient conditions. Both SIMPAK and MCOMP have subroutines that compute mixture physical property information as needed in the model calculation.

Description of the performance of a packed column requires both transfer coefficients and interfacial areas for mass and heat transfer. These quantities, along with column hydraulics, will determine packing effectiveness.

Table 1. *Components Included in Model*

Butane	*Ethylene*	*Methanol*
Carbon Dioxide	*Hydrogen*	*Nitrogen*
Carbon Monoxide	*Hydrogen Sulfide*	*Propane*
Carbonyl Sulfide	*Methane*	*Propylene*
Ethane		

Table 2. Information Input To MCOMP (Corresponding to Conditions of Integrated Run AMI-53/GO-73)

Gas Mass Vel. mol/hr/ft²	Gas Temp. In °F	Liq. Mass Vel. mol/hr/ft²	Liq. Temp. In °F
17.88	42.53	127.46	-4.28

Pressure Atmos.	The Key Comp.	Fract Key Comp. Absorbed
17.890	1	0.99900

Comp. No.	Comp.	Inlet Gas Mol. Fract	Inlet Liquid Mol. Fract
1	CO_2	0.2213	0.0000
2	H_2S	0.0017	0.0000
3	COS	0.0001	0.0000
4	MEOH	0.0000	1.0000
5	H_2	0.3292	0.0000
6	CO	0.1724	0.0000
7	N_2	0.2112	0.0000
8	CH_4	0.0529	0.0000
9	C_2H_4	0.0045	0.0000
10	C_2H_6	0.0065	0.0000
11	C_3H_6	0.0000	0.0000
12	C_3H_8	0.0000	0.0000
13	C_4H_{10}	0.0000	0.0000

Binary Gas Diffusivity Option:	Wilke-Lee Modification to Chapman-Enskog
Binary Liquid Diffusivity Option:	Wilke-Chang Modification to Stokes-Einstein Equation
Mixture Liquid Diffusivity Option:	Modification of Holmes et al. to Wilke-Chang Equation
Transfer Coefficient Option:	Onda et al.
Packing Height	7.10 ft
Packing Type	¼-in. ceramic Intalox saddles
Packing Increments	10.0
Interface Area Coor.	1.00
Maximum Iterations	0
Convergence Tolerance	0.0000100 0.0001000 0.0150000 0.0050000
Convergence Accel.	0.50000

Although the literature contains a great deal of information about the performance of certain packings with particular systems, prediction of the performance of packings for which data are not available is difficult at best.

Mass transfer data, obtained from a particular column and system, are often reported as the product of the transfer coefficient and the interfacial area because it is often difficult, if not impossible, to separate the two quantities. However, using data obtained on one system to predict mass transfer rates in another often requires estimation of both quantities.

While many investigators have studied the effectiveness of various packings, few have tried to generalize their results to other operating conditions and systems. For this reason, design methods for packed columns still rest to a large extent on experience and vendor advice. Large safety factors have to be used to ensure that the column meets design expectations.

To be able to predict column performance accurately, mass transfer coefficients in the liquid and gas phases must be known. If this information is not available for the system being used, correlations based on experimental data can be used to predict them. The accuracy of the correlations depends on the accuracy of the data used in their development. This is especially important because experimental difficulties make the measurement of mass transfer coefficients susceptible to significant error.

While it is relatively easy to determine the dry surface area associated with a particular type and size of packing, it is difficult to determine the amount of the packing surface that is effectively used in an irrigated section of packing. Several factors are important: the type of process for which the column is used influences the amount of surface area; the surface tension of the liquid passing through the packing determir.es the wetting characteristics; and the shape, size, and material used for packing.

Because there is no widely accepted correlation for either mass transfer coefficients or interfacial area in packed columns, four options have been included in SIMPAK and MCOMP. The option to be used in a particular computation is chosen in the program input.

Experimental Method

Details of the gasifier and the entire pilot plant are provided in Volume I. Data required to compare experimental results with model predictions were generated in two ways: (1) syngas runs involved the use of a gas-mixing manifold where gas was fed from bottles and metered to the absorber through a flow controller; and (2) integrated runs involved the use of the fluidized bed gasifier to manufacture gas to be fed to the absorber. The procedures used to operate the absorber and the rest of the acid gas removal system were similar in both types of runs.

After steady state was achieved, gas was sampled at the inlet to the absorber (sour gas) and at the outlets from the absorber (sweet gas), the flash tank (flash gas), and the stripper (acid gas). After this first sampling period, the system was operated for at least an hour before a second set of samples was taken. After the second set of samples, the system was usually shut down and the run concluded.

After conclusion of the run, collected gas samples were analyzed and used to check mass balances around the entire AGR system, comprised of the absorber, flash tank, and stripper. Results from these mass balances were then used to assess the quality of the run. Deviations of more than 10% for either the overall balance or the individual component balances usually resulted in disallowing the results of the run. Some judgment was used, however, in analyzing the results, especially if the reason for the discrepancy in the material balance was known. Also, for several components fed to the system at concentrations less than 1 mole %, mass balance deviations of greater than 10% may have resulted in disallowing the run results. As a rule, however, the mass balance results from a particular run met the given criteria. The mass balance results for both the syngas and integrated runs are included in Volume II.

Discussion of Results

Comparison of Experimental Data to SIMPAK

In general, model predictions compared well with experimental data. The experimental results from syngas Run AM-50, typical of the better runs in this series, are

illustrated in Figure 3. It shows excellent agreement between the computed liquid temperature profile and the experimental data.

Agreement between measured syngas temperature profiles and calculated values showed that the model satisfactorily predicted column performance. The plots of the height of packing versus the liquid temperature show that the shape of the predicted curve closely follows the experimental data. Uncertainties in temperature measurement probably account for the infrequent differences between predicted and experimental results.

Table 3 compares predicted and measured temperatures of liquid leaving the absorber for completed runs satisfying the closure requirements on mass balances. That these compare closely confirms the accuracy of the flow meter calibrations, the sampling and analytical techniques, and the validity of the assumptions made in estimating the heat of solution of CO_2 in methanol from Henry's Law constants.

Comparison of Experimental Data to MCOMP

Using the method established for comparing results from SYNGAS runs to the prediction from SIMPAK, results from integrated runs were compared to predictions from MCOMP. For each integrated run, the computed liquid temperature profile was compared to the experimental data. The measured sweet gas compositions were also compared to predicted results. Results are shown in Table 4, which compares predicted and measured outlet liquid temperatures. Agreement between these two sets of values is an indication of the accuracy of the adiabatic assumption as well as a check on sampling and analytical techniques. In all cases, the correlation of K. Onda et al. was used to estimate the mass transfer coefficient and interfacial area. Because MCOMP handles up to 13 components, considerably more computation time was required than was the case with SIMPAK. For this reason, 10 packing increments were chosen to economize on computer time.

In all the comparisons of model predictions to experimental results, CO_2 was the key component. While any of the components in the model could be used as the key component, the absorption of CO_2 could be followed most easily through the column liquid temperature profile. The other components were either sparingly soluble in methanol or

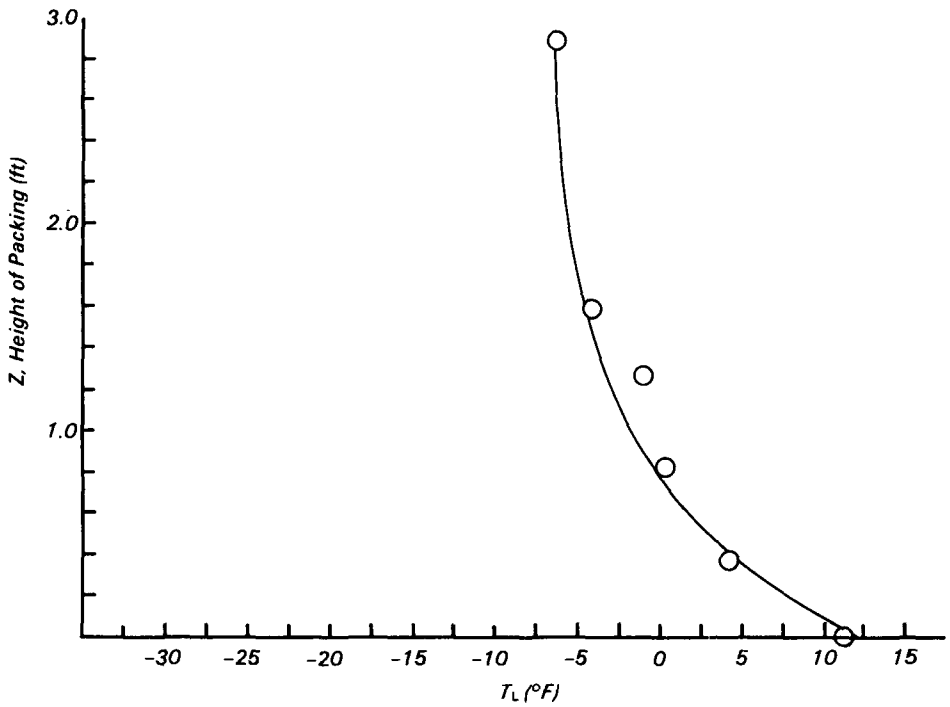


Figure 3. Comparison of data from Syngas run AM-50 with model calculations using the Onda correlation and 50 packing increments.

Table 3. Comparison of Predicted and Experimental Outlet Liquid Temperatures (T_L)

Run	T_L In, °F	T_L Out, °F	Computed T_L Out, °F
24	-33.69	6.36	6.91
25	-34.02	3.73	1.53
26	-33.30	9.68	9.33
27	-28.55	14.41	—
32	-31.79	11.34	9.34
33	-28.91	5.78	6.78
34	-29.33	15.75	17.11
38	-18.62	10.70	7.69
50	- 6.33	11.17	12.03
55	- 4.17	29.01	32.18

Table 4. Comparison of Predicted and Experimental Outlet Liquid Temperatures (T_L) For Integrated Runs

AGRS Run No.	Gasifier Run No.	Mass Balance	T_L In °F	T_L Out °F	Computed T_L Out, °F
30	56	100.0	-28.27	4.83	5.47
35	59	102.4	-34.65	0.72	3.11
36	60	104.0	-27.75	-1.72	-0.84
37	61	103.3	-22.20	7.19	8.22
43	68B	103.8	-34.07	-4.53	-2.82
44	69	102.3	0.84	24.16	[a]
45	70	103.0	-34.07	-19.23	-19.74
47	71B	104.8	-33.07	-17.13	-17.35
52	72	101.4	-30.70	-15.11	-14.81
53	73	103.1	- 4.28	8.72	10.13
57	76	99.2	-32.70	-2.45	[a]
59	78	101.8	- 2.92	9.50	9.29
60	79	95.5	-16.00	5.98	2.44

[a]Runs failed to converge.

present in such small quantities that their absorption did not affect the measured temperature profile.

A series of integrated runs were made using gas produced from a devolatilized char to feed the absorber. The model predictions of MCOMP compared well with the actual liquid temperature profile and outlet gas composition for all runs. In these runs, the transfer of eight compounds (including the solvent) was considered. The difference between the actual and predicted H_2S and COS outlet gas concentrations was attributed to the presence of both compounds in the recirculated methanol fed to the absorber. This resulted from the inefficient stripping of these compounds from the rich methanol and led to a reduced driving force at the top of the absorber. Figure 4 uses data from run AMI-30/GO-56 to illustrate the comparison of model predictions with data from these runs.

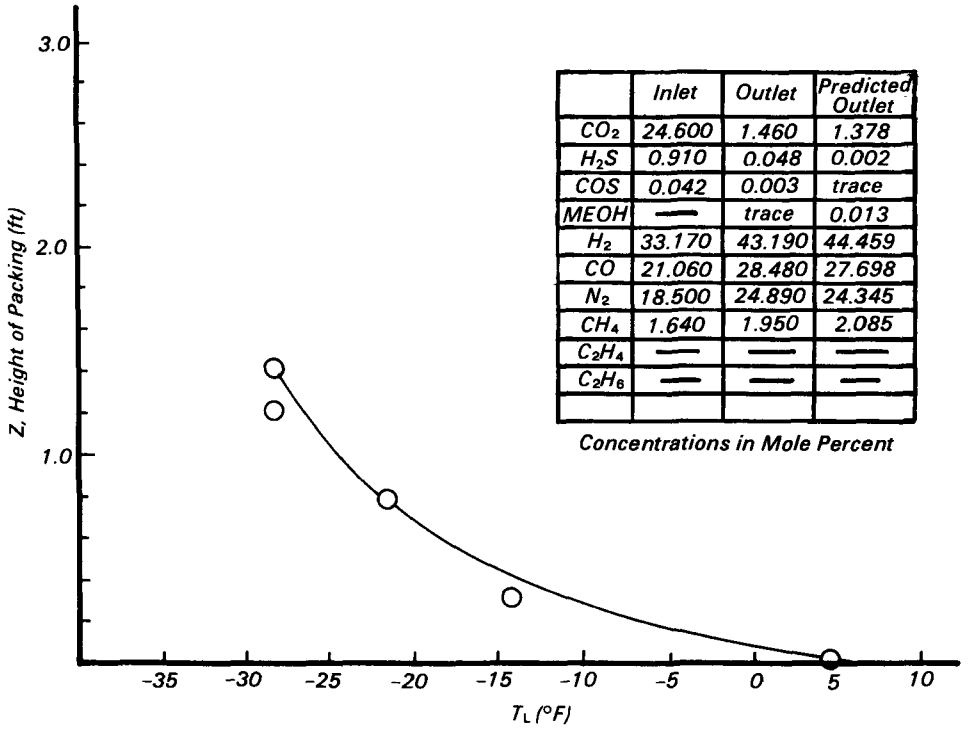
Another series of runs were made in the early 1980s, using a feed gas generated by the gasification of a New Mexico subbituminous coal. Gases produced from gasification of this coal were characterized by fairly low amounts of H_2S and COS, and relatively high levels of both aromatic and aliphatic hydrocarbons. Only compounds that could be analyzed with some accuracy and that were important in mass balance calculations were included in the computer model runs. An example of the comparison of model predictions to data obtained in these runs is given in Figure 5 for Run AMI-60/GO-79.

In general, the computer prediction compared well with the experimental data. Comparisons of predicted and experimental outlet liquid temperatures, and indications of the closure of mass balances for all integrated runs are given in Table 4.

Figures 6 and 7 show the variation in composition of the gases as they move through the absorber for AMI-53. Two graphs are presented because of the range of compositions encountered; note that the acid gases H_2S, COS, and CO_2 are readily absorbed, but that the product gases CO, H_2, and CH_4 increase in concentration through the column, mainly due to the concentrating effect of removing large amounts of the acid gases.

Minimum Liquid/Gas Ratio

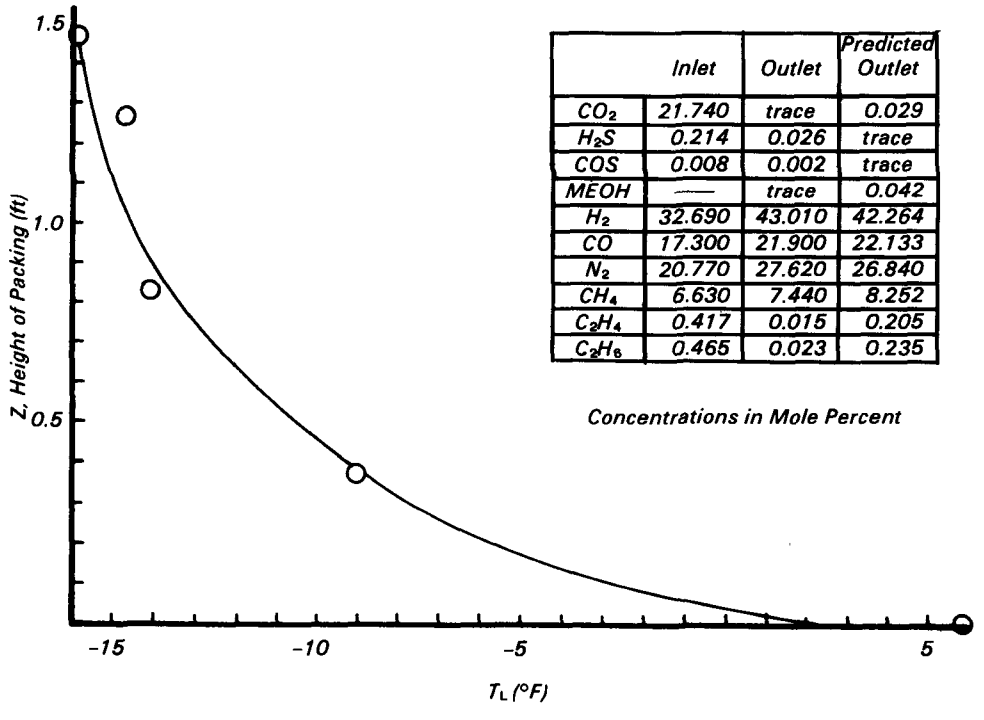
Specification of the liquid flow rate is required for absorber design. The minimum liquid/gas ratio, evaluated from



	Inlet	Outlet	Predicted Outlet
CO_2	24.600	1.460	1.378
H_2S	0.910	0.048	0.002
COS	0.042	0.003	trace
MEOH	—	trace	0.013
H_2	33.170	43.190	44.459
CO	21.060	28.480	27.698
N_2	18.500	24.890	24.345
CH_4	1.640	1.950	2.085
C_2H_4	—	—	—
C_2H_6	—	—	—

Concentrations in Mole Percent

Figure 4. Comparison of data from integrated run AM-30/GO-56 with model calculations using the Onda correlation and 10 packing increments.



	Inlet	Outlet	Predicted Outlet
CO_2	21.740	trace	0.029
H_2S	0.214	0.026	trace
COS	0.008	0.002	trace
MEOH	—	trace	0.042
H_2	32.690	43.010	42.264
CO	17.300	21.900	22.133
N_2	20.770	27.620	26.840
CH_4	6.630	7.440	8.252
C_2H_4	0.417	0.015	0.205
C_2H_6	0.465	0.023	0.235

Concentrations in Mole Percent

Figure 5. Comparison of data from integrated run AMI-60/GO-79 with model calculations using the Onda correlation and 10 packing increments.

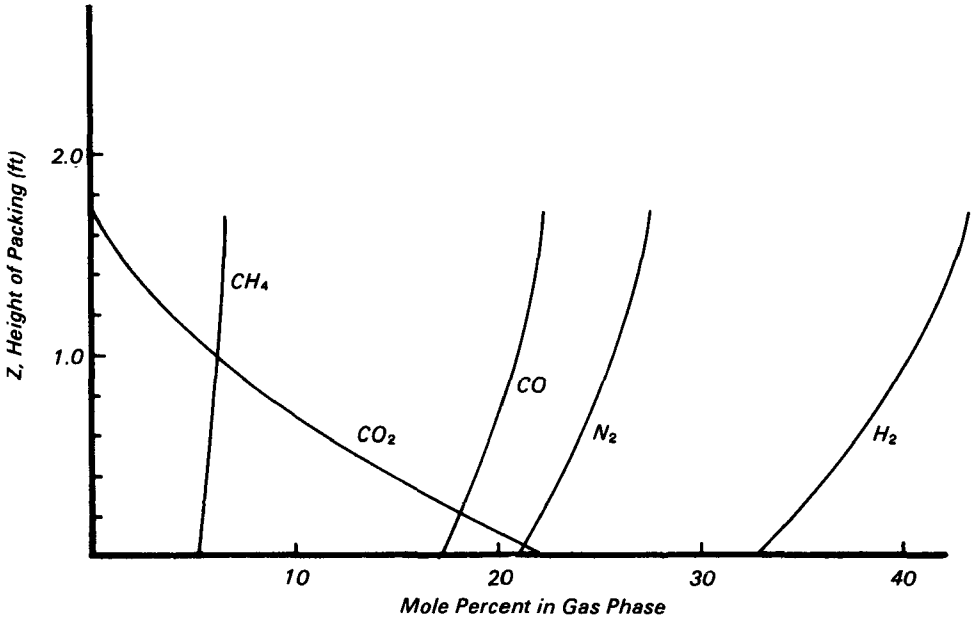


Figure 6. Calculated gas composition profile for conditions corresponding to integrated run AMI-53/GO-73.

physical property data, provides a useful limit in setting design flow rates.

SIMPAK was modified to estimate the minimum liquid rate for all syngas and integrated runs made for this study. The subroutine MINLG, included in SIMPAK, systematically brackets the minimum liquid rate until it can be determined to within 0.5 lb moles/hr/ft^2.

Comparison of Transfer Coefficient Correlations

Correlations of K. Onda et al. were used to estimate mass transfer coefficients and interfacial area in the computer runs comparing model predictions to experimental results. The correlations were chosen because they were developed with data obtained for small packing sizes and for cases where organic liquids, including methanol, were used. Earlier results showed that these correlations yielded excellent agreement between predicted and experimental data.

The following points can be made concerning other correlations of transfer coefficients that were tested in this study. The approach taken by W. R. Bolles and J. R. Fair seems to be the most sound: it correlates the product of the transfer coefficient and the interfacial area. However, Bolles and Fair suggested that the scatter in the data used to develop the correlation requires a safety factor of 1.70. They also suggested using a safety factor of 2.23 with the Onda correlation.

Results of this study indicate significant differences between the results of various researchers in developing generalized correlations for mass transfer in packed columns. This is partly the result of the scatter in the data used to develop these correlations. While Fair's approach avoids the problem of correlating the interfacial area and transfer coefficient separately, an effective generalized correlation for each quantity would be more desirable so that these results can be extended to a wider variety of conditions. At this point, it appears that additional experimental data and better methods of correlation would improve design procedures for packed columns.

Recommentations for Application of Model

The mathematical model of the packed absorber described here has been used to analyze the results of the experimental runs made for this study. Other objectives in developing this model included its use to extend the experimental results to situations not attainable in pilot plant operation. Future work will extend the packed absorber model to stripper operation and culminate in the development of a simulation package to describe the performance of the entire system.

A valuable use of a computer model is to evaluate the effect of changes in process variables on the final column design. This may require a complex optimization procedure or may be fairly simple if only one variable is involved. In either case, the computer model becomes the tool for the evaluation.

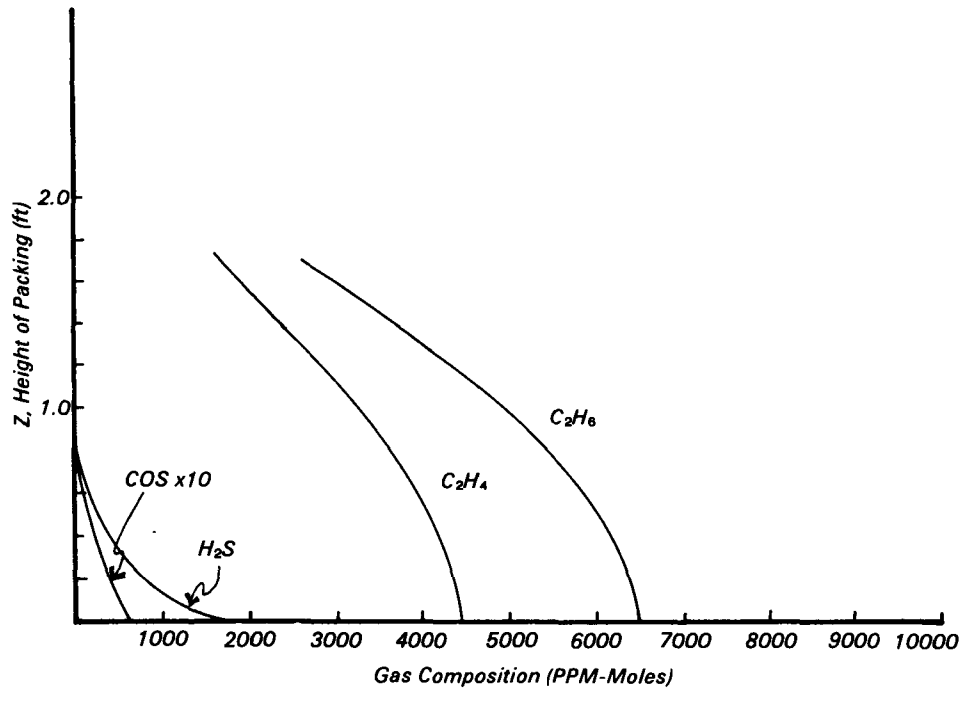


Figure 7. Calculated gas composition profile for conditions corresponding to integrated run AMI-53/GO-73.

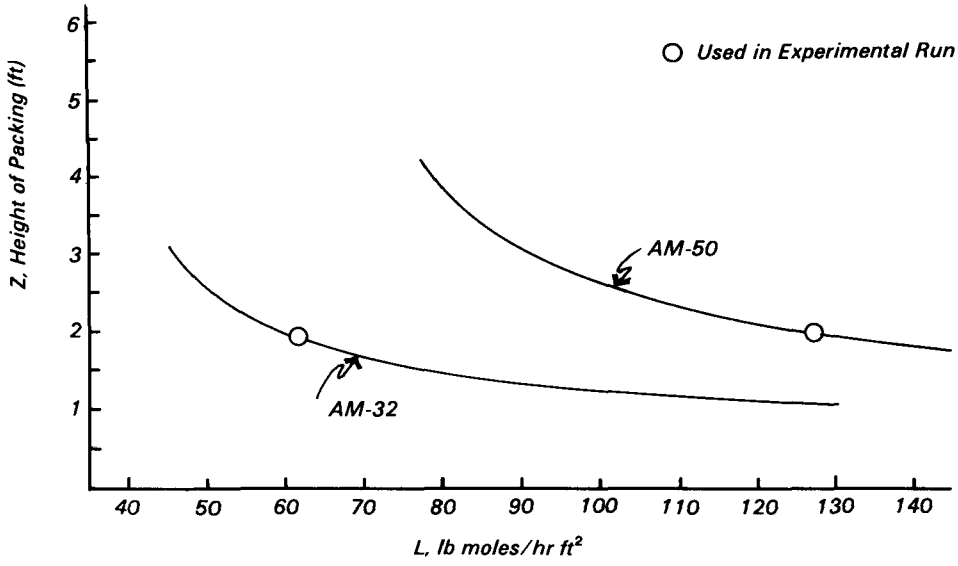


Figure 8. *Effect of Solvent Flow Rate on Packing Requirements for conditions corresponding to Syngass runs AM-32 and AM-50.*

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***N. Dean Smith** is the EPA Project Officer (see below).*

The complete report, entitled "Coal Gasification/Gas Cleanup Test Facility: Volume IV. A Mathematical Model of the Packed Column Acid Gas Absorber," (Order No. PB 84-113 083; Cost: $17.50, subject to change) will be available only from:

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Syngas Runs AM-32 and AM-50 were used as base cases for SIMPAK to illustrate the use of the models developed here in a parametric study. Figure 8 shows the application of these models to examine the effect of liquid flow rate on required packing height. This kind of analysis can be performed with any of the specified operating variables in the process. That AM-50 was made at a higher inlet liquid temperature is partly responsible for the greater sensitivity of the packing height to the liquid flow rate. The solubility of CO_2 in methanol at the inlet liquid temperature of AM-50 is much less than for AM-32.

Nomenclature

b	total liquid holdup in column
b_j	holdup of j in liquid
G	total gas molar flow rate
G_j	gas flow rate of j
H_b	specific enthalpy of liquid holdup
H_{ex}	enthalpy loss to surroundings
H_G	specific enthalpy of gas
H_L	specific enthalpy of liquid
j	chemical species j
L	total liquid molar flow rate
L_j	liquid flow rate of j
t	time
T_L	liquid temperature
x_j	mole fraction of j in liquid
y_j	mole fraction of j in gas
z	height of packing

Conversion Factors

Readers more familiar with metric units may use the following factors to convert to metric equivalents:

Non-metric	*Times*	*Yields metric*
°F	*5/9(°F-32)*	*°C*
ft	*30.48*	*cm*
ft²	*0.09*	*m²*
in.	*2.54*	*cm*
lb.	*0.45*	*kg*

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