Condensation of Vapor Mixtures. 2. Comparison with Experiment

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The simple one-dimensional models of multicomponent condensation described in part 1 are used to simulate the performance of a wetted-wall column in which a gas/vapor mixture containing acetone, benzene, and nitrogen or helium is contacted with a liquid film containing only the first two of these components and a vertical tube condensing isopropyl alcohol and water from a vapor which also contains nitrogen or freon 12. The results obtained with the nonequilibrium models are in good agreement with the experimental data. The models which account for interaction effects (mass and molar reference frame) are in better agreement with the data than is the simple effective diffusivity method.

Introduction

There is a great shortage of experimental data on mass transfer in multicomponent vapor(plus inert gas)–liquid systems. Most of the published works deal with absorption (or condensation or evaporation) of a single species in the presence of a nontransferring component. A set of ternary mass-transfer experiments was carried out by Toor and Sebulsky (1961) and Modine (1963) in a wetted-wall column and also in a packed column. These authors measured the simultaneous rates of transfer between a vapor–gas mixture containing acetone, benzene, and nitrogen or helium and a binary liquid mixture of acetone and benzene. Vapor and liquid streams were in cocurrent flow in the wetted-wall column and in countercurrent flow in the packed column. Their experimental results show that diffusional interaction effects were significant in the vapor phase especially for the runs with helium as the inert gas. The theoretical model used by Toor and Sebulsky and Modine to explain their experimental results was based on the generalized driving force approach of Toor (1957). More recently, Krishna (1979a, 1981) used the wetted-wall column data of Modine to test the applicability of the Krishna–Standart (1976) multicomponent film model and also the linearized theory of Toor (1964) and Stewart and Prober (1964).

Webb and Sardesai (1981) report the results of a number of experiments involving the condensation of isopropyl alcohol and water in the presence of nitrogen or freon 12 as noncondensing gas. Webb (1982) reviews other data obtained by his co-workers, including the results of Deo (1981) obtained in a condenser of annular geometry and Shah (1981) who condensed water and methanol in the presence of a variety of inert gases in a condenser consisting of 50 horizontal tubes mounted in a rectangular duct. Numerical simulations of these experiments are discussed by Webb and Sardesai (1981) and Webb (1982) (who used the Krishna–Standart, Toor–Stewart–Prober, and effective diffusivity methods to calculate the condensation rates) and by McNaught (1983) (who also used the equilibrium model of Silver (1947)).

In this paper, we use the data of Modine (1963) and of Webb and Sardesai (1981) to test the applicability of the turbulent eddy diffusivity models and the computational procedures developed in part 1. This is the first work to compare the turbulent eddy diffusivity models to the results of experiments.

Experiments

Modine (1963) carried out his experiments in an adiabatic wetted-wall column 0.6096 m in length and 0.025 019 m inside diameter. A gas–vapor stream containing acetone (1), benzene (2), and nitrogen (3) or helium (3) was contacted with a cocurrently flowing liquid stream containing only the first two components. Seven runs were carried out with nitrogen as the inert gas and six runs with helium. The experimental results are most accessible in a paper by Krishna (1981); for this reason, they are not repeated here. It is worth noting, however, that most of the experiments involve the simultaneous evaporation of benzene and condensation of acetone. The experiments cover a rather narrow range of liquid compositions (mole fraction acetone about 0.1) and vapor flow rates (Reynolds numbers around 9000).

The most comprehensive set of experiments of binary vapor condensation in the presence of an inert gas has been carried out by Webb and co-workers (1981, 1982). The experiments of Sardesai (1979) were carried out in a vertical condenser tube 1 m long and 0.023 m internal diameter. Eight runs were carried out in which isopropyl alcohol (1) and water (2) were condensed in the presence of nitrogen as the inert gas and another seven runs with freon 12 as the noncondensable component. These experiments cover a somewhat larger range of Reynolds numbers (7000–20 000) and vapor composition. Complete details of the experiments are available in the thesis of Sardesai (1979); a summary of the results (sufficient for our purposes) is given by Webb and Sardesai (1981).

Simulations

Our simulations of Modine's experiments were purposely made as similar as possible to those of Krishna (1981) so that we can more easily compare our results with his. For these simulations, the column was divided into 24 sections, each of which was modeled by a nonequilibrium section as described in part 1. Physical properties like density, viscosity, heat capacity, and thermal conductivity were evaluated by using the same methods employed by Krishna (1981). The Fanning friction factor (needed in

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the calculation of the heat- and mass-transfer coefficients) was estimated by using an expression obtained by Modine in his column (see Krishna (1981) for details). K values were estimated by using the Antoine equation for the saturation vapor pressures and the Wilson equation for the liquid-phase activity coefficients. Latent heats of vaporization were calculated by using the Watson equation (Reid et al., 1977). All physical and thermodynamic properties were evaluated separately in each section of the condenser. In this way, we were able to reproduce exactly the results quoted by Krishna (1981) (see, also, Krishna (1975) for more detail.)

Webb and Sardesai used a fourth order Runge-Kutta method to integrate the differential equations modeling the condenser (see part 1). For our simulations of Sardesai's experiments, the condenser was divided into 40 sections. By and large, the methods used to calculate physical properties were the same as those used by Webb and Sardesai in their own simulations (see Sardesai, 1979). The Fanning friction factor was calculated from a correlation obtained by Webb and Sardesai from dry gas cooling experiments carried out in in their condenser. Our simulations of Sardesai's experiments were made easier by employing the measured wall temperature profile as described in part 1. This allows us to dispense with the energy balance for the coolant and the uncertainties in the estimation of the coolant heat-transfer coefficient (the dimensions of the annular space occupied by the coolant and the coolant flow rate are not specified in the paper by Webb and Sardesai (1981)). The wall temperature at positions located between the thermocouples was estimated by linear interpolation.

Our initial objective was to assess the ability of the turbulent eddy diffusivity model and the other interactive models to simulate the experiments of Modine and Sardesai. The simulations of both sets of experiments were carried out by using a number of different models of vapor-phase mass transfer: (1) the Krishna–Standart (1976) method, (2) the linearized theory of Toor (1964) and of Stewart and Prober (1964) in the molar frame of reference, (3) the explicit method of Krishna (1979b), (4) the explicit method of Taylor and Smith (1982), (5) the turbulent eddy diffusivity model of Krishna (1982), (6) the linearized theory in the mass frame of reference, and (7) an effective diffusivity method.

The first four of these methods yielded results that were identical within four significant figures. Henceforth, we shall not try to distinguish these methods and refer to them collectively as the "molar reference frame" (or, more simply, molar frame) models (all four methods are based on the assumption of constant molar density—see part 1 for further discussion). We also found that methods 5 and 6 could not be told apart, at least on the scale of Figures 1–5; we shall refer to these two methods as the mass reference frame or mass frame models (since they are based on the assumption of constant mass density—see, again, part 1 for further discussion). Method 7 is the only one which does not attempt to account for diffusional interaction effects in the vapor phase.

Subsequently, we investigated the sensitivity of the model predictions to the liquid-phase mass-transfer coefficient. Note that the liquid (or condensate) film contains only two species in both sets of experiments. This makes the calculation of a mass-transfer coefficient particularly simple (see, again, part 1).

Results and Discussion

The results of our simulations of Modine's experiments are shown in Figures 1 and 2. The first figure provides a comparison between the mass-transfer rates measured by Modine (1963) and the rates predicted by using the three different classes of models for the vapor-phase mass-transfer process: the molar frame models, the mass frame models, and the effective diffusivity models. The results for the molar frame models and for the effective diffusivity model agree exactly with the results given by Krishna (1981). It is clear from this figure that the models that account for interaction effects are in better agreement with the experimental data than is the effective diffusivity method. Overall, the mass frame models (turbulent eddy diffusivity and Chilton–Colburn) do a little better than the molar frame models. It is interesting to observe that although all the interactive models (mass and molar frame) predicted that acetone experienced reverse mass transfer in run 7 (helium as inert gas), in agreement with the experimental findings (Krishna, 1981), the mass frame models were the only ones which correctly predicted the observed net evaporation of acetone.

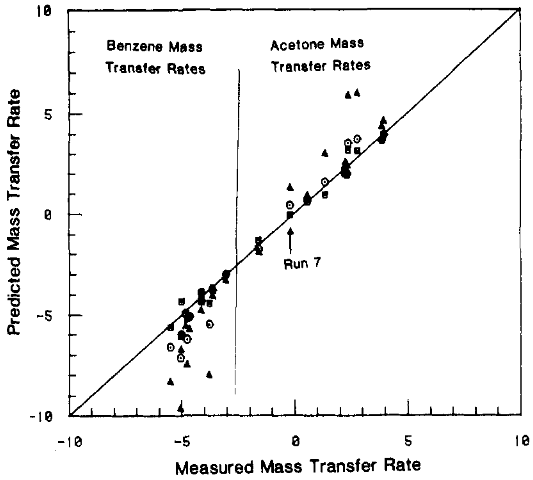

Figure 1. Comparison of predicted and measured mass-transfer rates. Experiments by Modine involving mass transfer between a falling liquid film of acetone and benzene and a gas–vapor mixture containing, in addition to these two components, nitrogen or helium. Finite rate model for the liquid phase.

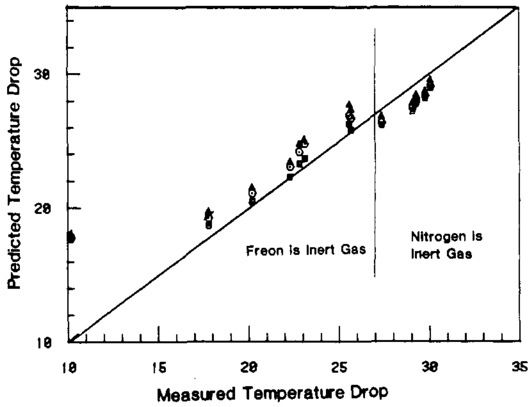

Figure 2. Comparison of predicted and measured temperature drop over a vertical tube condensing isopropyl alcohol and water in the presence of nitrogen or freon 12. Experiments by Webb and Sardesai.

Simply by varying the numerical value of the liquid-phase mass-transfer coefficient, it is possible to approach either of the two extremes of liquid mixing; complete mixing with infinite (or very high) liquid film mass-transfer coefficient or no mixing at all with a zero liquid film mass-transfer coefficient. Figure 1 was obtained with the (non-zero, noninfinite) value of the liquid-phase mass-transfer coefficient used by Krishna (1981). The no-mixing

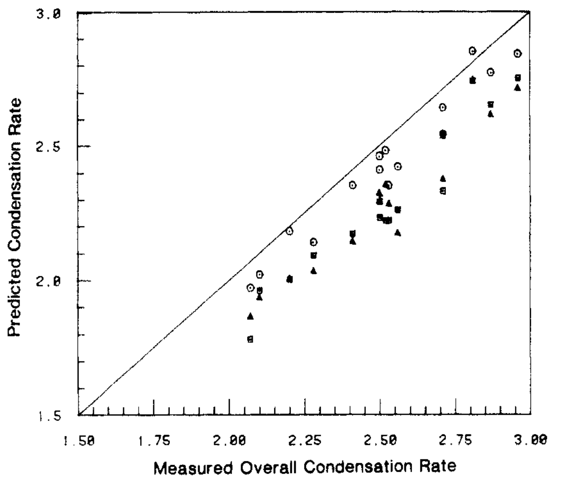

Figure 3. Comparison of predicted and measured overall rates of condensation of isopropyl alcohol and water in the presence of nitrogen or freon 12. Experiments of Webb and Sardesai.

limiting case is completely unable to model Modine's experiments. For this special case, the flux ratios N_i/N_t must be equal to the interface liquid composition, x_{iI}, which implies that each species transfers in the same direction, a circumstance certainly not encountered in Modine's experiments. The complete mixing limit, on the other hand, is very successful, at least when combined with the mass frame models. Figure 2 presents a comparison between the measured mass-transfer rates and those predicted by using the three models used to obtain Figure 1. The only difference between the calculations used to obtain Figures 1 and 2 is that the liquid-phase mass-transfer coefficient is 10 000 times higher for Figure 2 than for Figure 1. Further increase in this value did not change the results. The agreement between measurement and model prediction is, again, better with the mass frame models (better even than in Figure 1), while the molar frame models show a surprising decline in performance in the runs with helium as inert gas (for reasons that are not entirely clear to us). The effective diffusivity model gave the poorest predictions of the mass-transfer rates.

The results of our simulations of Sardesai's experiments are summarized in Figure 3 (predicted vs. measured overall temperature drop) and Figure 4 (predicted vs. measured total condensation rate). In contrast to the simulations of Modine's experiments, both extremes of condensate mixing yield almost identical results; those shown in Figures 3 and 4 were obtained by using the no-mixing option. It can be seen that the effective diffusivity methods give good predictions of the overall temperature drops (Figure 3) although there is little to distinguish any of the models here. We found that the Chilton–Colburn analogy consistently underpredicted the total condensation rates. This is a well-known drawback of the Chilton–Colburn analogy (Sherwood et al., 1975). Thus, we also investigated the use of the Gilliland–Sherwood correlation (Sherwood et al., 1975) for estimating only the mass-transfer coefficients for the molar frame models (1–4, 7) (heat-transfer coefficients were still calculated from the analogy). Overall, the best agreement between measurement and prediction was obtained with this combination of methods, and Figures 3 and 4 show these results. All three classes of model still tend to underpredict the total condensation rates (but not as much as was found by using the Chilton–Colburn analogy to estimate the mass-transfer coefficients), the molar frame models are a little better here than the mass frame models, and both classes of interactive models are quite a lot better than the effective diffusivity model.

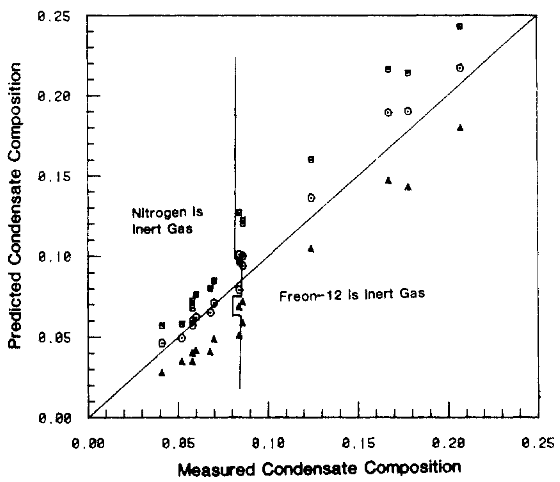

Figure 4. Comparison of predicted and measured mole fractions of isopropyl alcohol in a condensate of IPA and water. Experiments of Webb and Sardesai. Key to Figures 1–4: (⊙) molar frame models (Krishna–Standart, Toor–Stewart–Prober, explicit); (□) mass frame models (Chilton–Colburn and turbulent eddy diffusivity); (△) effective diffusivity method.

The condensate composition is a good indicator of the individual condensation rates; Figure 4 compares the measured and predicted condensate composition. Here, also, the molar frame models are superior to the mass frame models (which overpredict the rate of condensation of isopropyl alcohol in every case), and both types of interactive model are better than the effective diffusivity method (which underpredicts the rate of condensation of isopropyl alcohol in every case).

The simulations of Sardesai's experiments were surprisingly sensitive to changes in the wall temperature profile. The wall temperature varied over a few degrees in each experiment, and the results shown in Figures 3 and 4 were obtained by using the wall temperature profile reported by Webb and Sardesai (1981). As already noted, linear interpolation was used to estimate the wall temperature at locations between the thermocouples. The use of a wall temperature profile held constant at either the high, the low, or some average of the measured values was not able to predict the performance of the experimental condenser with anywhere near the same degree of success. This sensitivity emphasizes the need for accurate experimental measurements of the wall temperature profile. Incidentally, our use of the measured wall temperature profile (in place of the coolant energy balance, see part 1) meant that we were able to obtain even better agreement between model prediction and experimental measurement than was obtained by Webb and Sardesai (1981) in their simulations of their own experiments.

Finally, we quantify the magnitude of the discrepancies between prediction and measurement in Table I. We have chosen to report the percentage discrepancies for the condensation rates of isopropyl alcohol and water rather than for the total condensation rate and the condensate composition. This is done in order that the simulations of all experiments be quantified on the same basis. In view of our earlier remarks, there are no real surprises here; interestingly enough, at least one method predicts mass-transfer rates to an average discrepancy of 10% or less. Run 7 of the simulations of Modine's experiments is particularly sensitive to the mass-transfer model used. This is because acetone experiences reverse mass transfer in this run, and the overall transfer rate of acetone is an order of magnitude less than that of benzene. For this reason, the discrepancies for run 7 are reported separately in Table I. Note also that two data points have been

Table I. Average Deviations between Model Prediction and Experimental Measurement [Percentage Difference = (Predicted Quantity − Measured Quantity) × 100/Measured Quantity]

	mass-transfer model[a]			
quantity	1	2	3	4
Simulations of Modine's Experiments				
finite mass-transfer rate in liq phase				
acetone (av of 10 runs)	15.8	15.7	16.9	44.1
acetone (run 7)	−304.8	−14.6	−48.9	−637.8
benzene (av of 11 runs)	16.5	7.2	8.1	36.4
completely mixed liq				
acetone (av of 10 runs)	24.3	15.0	16.7	64.5
acetone (run 7)	−370.7	−31.1	−70.8	−759.8
benzene (av of 11 runs)	18.9	7.8	10.0	38.2
Simulations of Experiments by Webb and Sardesai				
isopropyl alcohol (av of 15 runs)	7.9	24.5	23.2	32.8
water (av of 15 runs)	2.3	4.9	5.7	4.8
temp drop (13 runs)	8.6	8.2	8.5	11.3

[a] Mass-transfer models: (1) molar frame models (see text), (2) mass frame Chilton–Colburn, (3) turbulent eddy diffusivity, (4) effective diffusivity.

omitted from the calculation of the temperature drop discrepancy; there is some disagreement between the tabulated data and a figure given in the paper by Webb and Sardesai (1981). In view of the possibility of a typographical error, we have ignored these points in the computation of the percentage deviations.

Concluding Remarks

The simple one-dimensional models of multicomponent condensation and cocurrent separation processes described in part 1 are well able to model the performance of a wetted-wall column operated by Modine (1963) and a vertical tube condenser operated by Sardesai (1979). The results obtained with the one-dimensional model are quite good enough for design purposes; it is doubtful if a more sophisticated boundary layer analysis could yield any better agreement.

As promised in part 1, we have provided experimental evidence of the superiority of mass-transfer models that account for interaction effects in the vapor phase. On the basis of the experiments simulated here, it can be seen that there are noticeable differences between the mass and molar frame models (more than was found in the simulations described in part 1), but it is not possible to clearly identify which of these models is the better. The mass frame models are better able to simulate Modine's experiments, whereas the molar frame models are better at simulating the condensation experiments of Sardesai.

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