CHAPTER VI

CONCLUSIONS

This work has provided significant advances towards the understanding of molecular diffusion by increasing the available data base for diffusion in liquid hydrocarbons at high temperatures, and by demonstrating the use of the rough hard sphere theory for prediction of diffusion coefficients. The first measurements of diffusion coefficients in an actual sample of Fischer-Tropsch wax have been obtained, and a method for estimating diffusion in Fischer-Tropsch wax has been suggested.

We have successfully constructed a Taylor dispersion apparatus capable of measuring diffusion coefficients at up to 600 K and 3500 kPa. The apparatus is the only known Taylor apparatus which can be used with liquid solvents which are solids at ambient conditions and is compatible with solvents which melt at temperatures up to 400 K. We have developed a new technique for measuring solvent density using the Taylor apparatus and have incorporated a capillary viscometer into the apparatus. During the course of the project, we also developed an improved method for analyzing Taylor dispersion peak data and a simplified technique for collecting gaseous diffusion data.

Using the Taylor apparatus, we performed well over 600 measurements of diffusion coefficients which were used to obtain the average values reported herein. Although not required for the project, we also measured solvent density at all experimental conditions, and viscosity for several of the solvents. Much of this additional data was not previously available, and allowed for the evaluation of several theories which relied on viscosity and density data. The density data which we collected ultimately led to the development of the rough hard sphere predictive equation which we have presented.

The rough hard sphere theory, which predicts a linear relationship between $D_{12}/\sqrt{T}$ and molar volume, has been extended for both dissolved gas and liquid solutes in alkane solvents over a wide range of temperatures. The hydrodynamic and activated state approaches to diffusion have been shown to be faulty. In addition to demonstrating that the form of RHS theory is correct, we have also demonstrated an equation for the a priori calculation of the diffusion coefficient. Unlike previous equations which require extensive solute and solvent property data, our model requires data for only one physical property, the solvent density. The model has been demonstrated to predict both gaseous and liquid solute diffusion in normal alkane solvents with carbon numbers ranging from 7 to 28 at temperatures to 570 K.

This work has provided the first general predictive equation for diffusion coefficients in liquids which is based on the rough hard sphere theory. Previous investigations have used RHS theory as an explanatory basis, but no attempts were made to generalize the results in the form of a simple predictive equation. We have also demonstrated that the RHS theory can be applied successfully to molecules which are unlikely to be spherical, such as n-octacosane.

Finally, we have provided measurements of diffusion coefficients for several important Fischer-Tropsch reactants and products in an actual sample of Fischer-Tropsch reactor wax at reactor conditions. We have shown that the diffusion coefficients in Fischer-Tropsch wax can be conservatively estimated by modelling the wax as a pure normal alkane with carbon number equal to the average carbon number of the wax. We hope that the success of this project will lead to further investigations of diffusion for other classes of compounds. Additional data is desperately needed before a complete theory of diffusion in complex mixtures such as Fischer-Tropsch wax can be developed.