CHAPTER 7

CONCLUSIONS

7.1 Basis Sets

The DZP quality basis set is the most appropriate for this work as it gave acceptable results within an appropriate time on available computers. The counterpoise correction was neglected, due to the tiny value of the BSSE and the great increase in computing time that would be required.

7.2 Potential Functions

In this work, all interactions of the particles in simulations are determined via the pair-potential functions, three-body NH_3-Ca(II)-NH_3 correction functions, and pseudopotential functions. Without the three-body correction, the simulation gave coordination numbers that were too high for the dilute solution. This indicates that three-body corrections are necessary for this system. In concentrated solutions, where electrons are delocalized in the solution, the pseudopotential model is used to represent the changes of the site-site interactions, leading to much less negative and positive interactions. It has been found that among the three pseudopotential models examined in the study, only one of the simulations, *the point ion pseudo-potential with the Gurskii dielectric function,* could be properly performed.

7.3 Structural and Dynamical Properties

The coordination number of NH_3-solvated calcium changed from 9 to 8 in dilute solution when three-body corrections were added, indicating the importance of three-body corrections.

In concentrated solutions, the solution structure is entirely changed. The ammonia molecules gather together to form irregularly shaped continuous clusters, and calcium ions float in the cavities. We hypothesize that free electrons are concentrated in these cavities. Calcium ions maintain a fixed separation distance of 4 Å inside the cavities, indicating metallic bonding.

The RDF and vibrational information obtained from simulations show agreement with experimental data, indicating that the potential functions employed are acceptable in describing the interatomic interactions.

7.4 Suggestions for Further Study

It would be interesting to further study the concentrated solutions of calcium in liquid ammonia. The following points can be investigated further for more accurate descriptions, knowledge, and understanding of the behavior of the solution.

- The ammonia-ammonia interaction may be re-assembled. Although the interaction employed in this work has been verified and works well in many studies, but the structure of the system has changed. It can be seen from the RDF and other data that the ammonia-ammonia distance and orientation are different from those of the dilute solution. However, it may be difficult to verify a new interaction function, because of the lack of necessary experimental data.
- It is know experimentally that structural and dynamical properties of the concentrated solutions are temperature and concentration dependent. To properly model such properties, a number of simulations with different pseudopotential models is required. In addition, experimental data at the molecular level are an importance key to justify reliability of the simulation models.