Abstract

There remain practical problems to predicting structures and properties of materials from first principles, though the foundation, quantum mechanics, has been established for many years. The goals of this research are to develop methods and tools that are accurate and practical, and apply them to important problems. Two aspects of the methodology are focused.

1. The development of accurate force fields based on \textit{ab initio} quantum mechanical calculations on prototype systems. Procedures were developed on polyvinyl chloride (PVC) and successfully applied on other types of polymers. They are very important to studying of amorphous polymers materials, for which current methods have not been useful in predicting important properties (e.g. moduli and glass temperature).

2. The development of Massive Parallel Simulation (MPSim) Software. MP-Sim is suitable for large systems (millions of atoms). It has the ability of including environmental variables (temperature, pressure, tension, and shear) and extracting physical properties (moduli and glass temperatures). The theories and algorithms implemented are summarized in the Appendix.

These methods and tools are applied to the accurate simulation of structures and properties of amorphous polymer materials and nano-materials.

Molecular dynamics (MD) simulation on polyethylene (chapter 6) was used to develop a general strategy for predicting glass transition temperatures which is expected to be very important in polymer industry. In chapter 7, these strategies were successfully applied to three important fluoro polymers.

Single-walled carbon nanotubes (SWNT), recently discovered but not very well characterized, is an interesting new class of materials. Using an accurate force field, structures and mechanical properties of these systems are studied. Chapter 2 shows that the dominating factor for deciding stable structures and mechanical properties
is the tube size, not chirality. The behavior of (10, 10) nano-tube under bending are studied (chapter 3) based on energy of hypothetical toroids with different radii. Yielding curvature of $1/R_s$ ($R_s = 183.3$ Å) where elastic bending becomes plastic response is found. In chapter 4, closest packing of $K_5C_{80}$ with the distribution of $K$ atoms along tube surface similar to the stacking of stage one $K_1C_8$ is established as the optimum structure of K-doped SWNT crystal.