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# The effect of empirical potential functions on modeling of amorphous carbon using molecular dynamics method

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ABSTRACT

Empirical potentials have a strong effect on the hybridization and structure of amorphous carbon and are of great importance in molecular dynamics (MD) simulations. In this work, amorphous carbon at densities ranging from 2.0 to 3.2 g/cm$^3$ was modeled by a liquid quenching method using Tersoff, 2nd REBO, and ReaxFF empirical potentials. The hybridization, structure and radial distribution function $G(r)$ of carbon atoms were analyzed as a function of the three potentials mentioned above. The ReaxFF potential is capable to model the change of the structure of amorphous carbon and MD results are in a good agreement with experimental results and density function theory (DFT) at low density of 2.6 g/cm$^3$ and below. The 2nd REBO potential can be used when amorphous carbon has a very low density of 2.4 g/cm$^3$ and below. Considering the computational efficiency, the Tersoff potential is recommended to model amorphous carbon at a high density of 2.6 g/cm$^3$ and above. In addition, the influence of the quenching time on the hybridization content obtained with the three potentials is discussed.



## 1. Introduction

Amorphous diamond-like carbon (DLC) films have been intensively studied theoretically and experimentally over the past few years due to their beneficial material properties such as high hardness, high wear resistance, low friction coefficient and high chemical inertness [1–5]. For instance, DLC films have been widely used as protective coatings against wear and corrosion in areas such as optical windows, hard disk drives and micro-electromechanical devices (MEMS) [6]. Such practical applications of DLC films require their ongoing investigation of the mechanical and tribological properties.

A number of studies can be found in the literature dealing with the mechanical and tribological properties of DLC films experimentally [7–10] and numerically [11,12]. Mohrbacher et al. [7] investigated the friction mechanism of DLC films by rubbing DLC with corundum, and found that the coefficient of friction (COF) decreased as the normal contact force increased. Also, it was found that DLC tends to graphitize under a large load, which can improve local frictional performance. Wu et al. [7] and Wang et al. [10] studied experimentally the friction mechanism of DLC and found that the contribution of aluminum and titanium atoms can improve mechanical properties of DLC.

It is very difficult to study the structure of amorphous carbon and conduct experimental studies of ultralow friction due to extremely low thickness of DLC and exclude substrate effects [3]. Hence, Molecular dynamics (MD) simulations can be used to investigate the tribological properties of amorphous carbon since the coordinates of all atoms can be calculated as a function of time [11,12]. Gao et al. [13] simulated the friction process between a pure diamond and DLC films to investigate the tribo-chemical reactions under a critical load. The results obtained by Gao were found in a good agreement with experimental results, and proved that the MD approach is a promising method to study the tribological properties of the amorphous carbon.

In MD simulations, a lot of theories, such as density-functional theory, tight-binding theory, and empirical potentials, have been applied to study the structural and mechanical properties of amorphous carbon [14]. Empirical potentials which have the advantage of high computational efficiency are commonly used to study mechanical properties of DLC. There are many types of empirical potentials used to model the interatomic interactions of carbon atoms, including Tersoff potential [15], Reactive Bond Order (REBO) potential [16,17], and its revised versions such as Reactive Force Field (ReaxFF) potential [18] and Environment Dependent Interaction potential (EDIP) [19] to name a few. The choice of empirical potential is one of the fundamental and important assumptions in

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**Table 1**
Number of carbon atoms at different densities.

| Density (g/cm$^3$) | 2.2 | 2.4 | 2.6 | 2.8 | 3.0 | 3.2 |
|---|---|---|---|---|---|---|
| Atom number | 882 | 962 | 1043 | 1122 | 1203 | 1282 |

the MD approach since it can affect the properties of amorphous carbon during the MD simulations.

In addition, it is important to determine the hybridization content and structure of the amorphous carbon to study its mechanical and tribological properties which strongly depend on the hybridization content and bond types of carbon atoms. There are two common methods to model the amorphous carbon in MD. The first one simulates an ion deposition processes and requires a careful modeling design and a significant computation time. This method is often used to explore the process of thin-film deposition from energetic species [20]. The second method simulates a quenching process of a molten carbon resulting in its solid phase state. Liquid quenching method is computationally efficient and achieves considerable success in reproducing experimentally observed properties of amorphous carbon [21]. No matter which methods are used, the choice of the empirical potentials has a significant influence on the calculation accuracy of the model. In this paper, liquid quenching method is used to model amorphous carbon.

As can be seen from the literature, no work can be found addressing the effect of empirical potentials on the properties of DLC. In this study we will focus on this topic and study the influence of the three types of potentials, specifically the Tersoff potential, the 2nd REBO potential and the ReaxFF potential on DLC parameters. These parameters include the sp$^3$ content as a function of density, the arrangement of the amorphous carbon atoms, and the radial distribution functions $G(r)$.

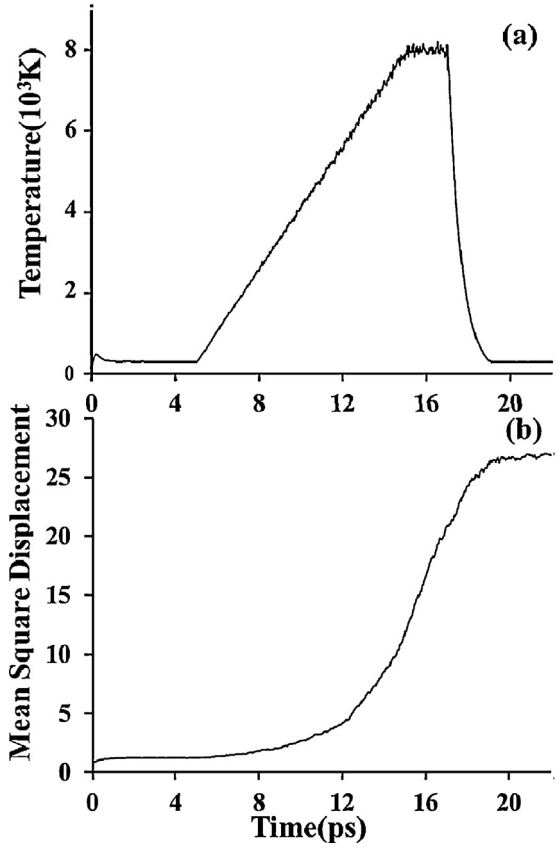


**Fig. 1.** (a) The temperature of the system as a function of time. (b) The mean square displacement (MSD) of the system as a function of time.

## 2. Empirical potential functions of carbon

The earliest computational method for amorphous carbon simulation was presented by Car and Parrinello [22], where the density functional theory (DFT) and molecular dynamics were combined together. However, this method was found to be computationally inefficient.

Tersoff developed a potential function for silicon in 1986 [23] and for carbon in 1988 [15]. It was the first computationally efficient potential function to study different hybridization states of carbon

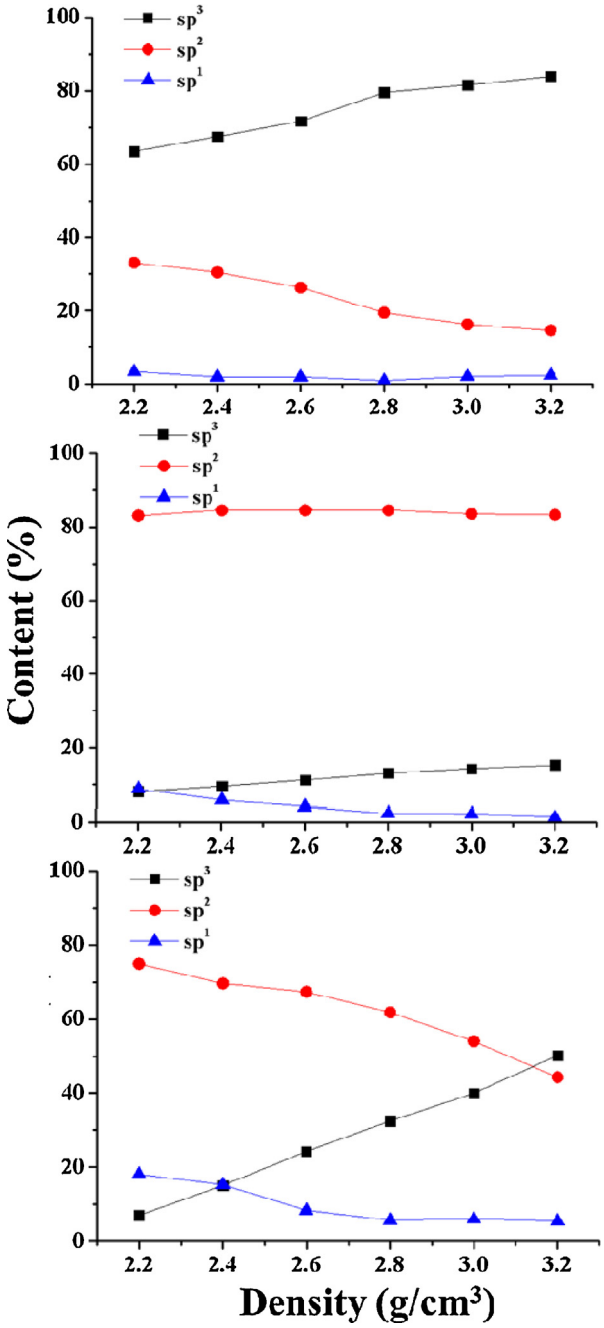


**Fig. 2.** Percentage of sp$^1$, sp$^2$, sp$^3$ hybridization states as a function of density of the amorphous carbon generated by (a) the Tersoff, (b) the 2nd REBO, and (c) the ReaxFF potentials.