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Microscopic Mechanism of Cellulose Bond Breaking and Bonding Based on Molecular Dynamics Simulation

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ABSTRACT The ReaxFF molecular dynamics simulation and Monte Carlo method were adopted to analyze the pyrolysis process of cellulose in insulating papers from the perspective of microscopic atom. Molecular dynamics failed to continuously describe the motion behavior of an atom. According to this principle, the system can only calculate the atomic state on the node and then move the atom by a time step to continue calculating the atom. This paper would put forward the optimal step setting method of cellulose thermal decomposition in insulating paper: setting one step every other 0.1 fs. Specifically, for small molecules with a simple structure, such as H_2O, the step size was set to 0.4 fs or less, while the step size of macromolecules with complex structures (CH_2O_2) should be set to 0.2 fs or less. In addition, the relationship between the step size and the temperature to which the system was heated was given as well in this paper. In previous literatures, empirical values were used to set the step size. This study would not only provide a theoretical basis for the study on the bond formation and fracture process of cellulose pyrolysis products, but also offer the data and guidance for related fields in the future, thus rendering an efficient simulation process.

INDEX TERMS Molecular dynamics, step size, cellulose, pyrolysis.

I. INTRODUCTION

Oil-immersed power transformer is the core equipment of power transmission and transformation system, whose safe and effective operation depends on multiple factors, such as service life, safety and aging [1]–[4]. The short circuit, lightning strike or partial discharge and other faults of transformer and its long-term operation would lead to the deterioration of electrical insulation and mechanical properties [5], [6]. The cellulosic insulating paper is a type of solid insulation material, which is widely used in electrical equipment such as wires and cables, transformers and capacitors [7], [8]. In the process of operation, the transformer insulation paper would have pyrolysis reaction under the joint action of multiple factors, such as local overheating in the transformer [9]–[11]. The insulating paper is mainly composed of cellulose [12]. The final products of cellulose pyrolysis at high temperatures are H_2O, CH_2O_2, CO, CO_2, methanol, ethanol, free radicals and small molecular acids [13]–[16]. Although the mechanical properties of insulating paper were reflected in its macroscopic mechanical behavior, such as plastic deformation, creep deformation and tensile strength, its microscopic mechanism remained largely elusive. Experimental methods were frequently used in the previous studies on the molecular simulation. It was found that the transformer oil contained methyl, ethyl, methoxy and hydroxyl according to the gas chromatography-mass spectrometry analysis of the liquid products of rapid pyrolysis of cellulose by Liao Yanfen and other scholars from Zhejiang University [17], [18]. Unfortunately, the macroscopic analysis methods used in

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the above studies failed to reveal the microscopic mechanism of cellulose pyrolysis or bond cleavage. It was worth noting that the molecular simulation method had attracted more and more attention in the in-depth exploration of the microscopic mechanism of cellulose pyrolysis. Hu Jian from Chongqing University utilized MaterialStudio software and COMPASS forcefield to simulate the pyrolysis process of cellulose [19]. However, traditional forcefields like the COMPASS cannot simulate chemical reactions at the atomic level. In recent years, the ReaxFF (reactive force field), which was able to reflect chemical bonding and breaking, was extensively applied to describe complex chemical reaction processes [20]–[24]. By applying the chemical calculation to electrical engineering and microscopic tools to explain macroscopic phenomena, ReaxFF-MD was able to clearly describe the structural changes of reactants and the bonding and breaking of molecules [21], [25]–[28]. In order to further study the microscopic mechanism of cellulose in ReaxFF, a molecular model of cellobiose was established in this paper. Combined with Monte Carlo, the molecular dynamics method of ReaxFF reaction was used to simulate the pyrolysis reaction. The simulation step size is set to 0.1 fs, 0.2 fs, 0.3 fs, 0.4 fs, 0.5 fs, 0.6 fs, 0.7 fs, 0.8 fs, 0.9 fs and 1 fs respectively. The target temperature for each system is set at 1900 K [29]. The optimal step size in different states was set in the process of chemical bonding and breaking [30].

II. CALCULATION PRINCIPLE

A. ReaxFF FILED CALCULATION

As for the ReaxFF model where the concept of atomic type in the classical force field was no longer applicable, there was no connectivity between the atoms [31]. Instead, the Bond Order (BO) between any two atoms was calculated to determine the connectivity at the current moment. In the simulation of reaction kinetics, the list of atomic connectivity was constantly updated with the breaking and formation of chemical bonds. Therefore, the core of the ReaxFF reaction force field is the expression of BO. On that basis, the interaction between atoms was defined as BO function, which can be divided into bond, angle, dihedral angle, conjugate, Coulomb, van der Waals and adjustment term [32]. Except for bond interactions, all parts of intramolecular energy were expressed by bond order [21], as shown in formula (1).

$$\begin{aligned} E_{system} = {} & E_{bond} + E_{lp} + E_{over} + E_{under} + E_{val} \\ & + E_{pen} + E_{coa} + E_{C2} + E_{triple} + E_{tors} \\ & + E_{conj} + E_{H-bond} + E_{vdwaals} + E_{coulomb} \end{aligned} \quad (1)$$

These partial energies include bond energy E_{bond}, Lone pair of electronic items E_{lp}, valence angle E_{coa}, E_{pen}, E_{val}, atom under/over coordination E_{under}, E_{over}, torsion angle E_{tors}, van der Waals force $E_{vdwaals}$, Four-body action E_{tors}, E_{conj}, Hydrogen bond E_{Hbond}, CorrectionE_{C2}, Coulomb force $E_{coulomb}$.

B. MONTE CARLO CALCULATION METHOD

Monte Carlo calculations refer to the count of random numbers in calculations [33]–[36]. Monte Carlo calculates the integral formula as follows:

① The coordinates of the N atoms in the system are randomly generated.

② Calculate the potential energy $U\left(\overrightarrow{r}^{N}\right)$ and function F of the configuration.

③ Calculating the Boltzmann factor $\exp\left(-\frac{U}{k_B T}\right)$.

④ The Boltzmann factor is gathered, and the function is multiplied by the Boltzmann factor and accumulated, and step ① is returned.

⑤ When the N_t cycle is calculated, the average F obtained is:

$$\langle F \rangle = \frac{\sum_{i=1}^{N_1} F_{\mathrm{i}} \cdot \exp\left[-U_i\left(r^{\rightarrow N}\right)/k_B T\right]}{\sum_{i=1}^{\mathrm{N}} \exp\left[-U_i\left(r^{\rightarrow N}\right)/k_B T\right]} \quad (2)$$

III. SIMULATION METHOD OF ReaxFF-MD

A. CONSTRUCTION OF CELLULOSE MODEL

At present, the oil-impregnated transformer insulation paper (plate) contained more than 90% oil, and the degree of polymerization (DP) of the new insulation paper varied between 1600 and 2000 [37]. During the operation of the transformer, the DP dropped due to normal or abnormal aging factors. It was impractical to directly simulate the pyrolysis reaction of cellulose chains with over 1000 DP [38]. Mazeau *et al.* reported that the amorphous model composed of cellulose chains with different lengths showed no significant difference in molecular conformation or physicochemical property [15]. To save the computation time, the cellobiose ($C_{12}H_{22}O_{11}$) was used as a model for it was a repeating unit of cellulose, and as shown in Figure. 2, the dynamics simulation calculation was performed for the molecular reaction. In the molecular model of cellobiose, white represents a hydrogen atom, red for an oxygen atom, and gray for a carbon atom.

B. SIMULATION DETAILS

In this paper, ADF software is used to simulate the pyrolysis process of cellobiose, a cellulose structural unit of transformer insulation paper. ADF software is a package for detailed and quantitative analysis of reactivity, structures, and various properties in the gas phase and in solution and for complicated and large-scale calculations on a large variety of systems [39]. As shown in Figure 1, ten initial amorphous cell models of the same size (30.5 nm × 30.5 nm × 30.5 nm) were constructed, each containing 35 cellobiose molecules with a density of 0.7008 g/cm^3. There was a great difference in the density between the nature cellulose and the initial amorphous cell models, thus the geometric structure and energy optimization were carried out to make the system in a stable state (the model density was closer to the actual density

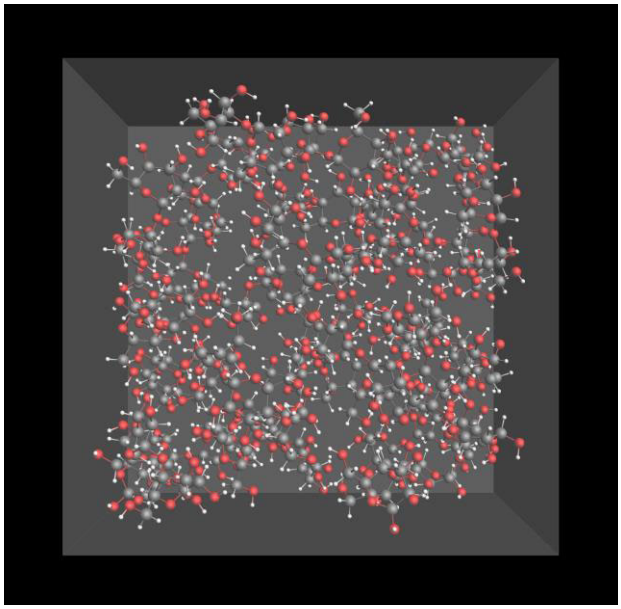

FIGURE 1. Initial amorphous cell model.

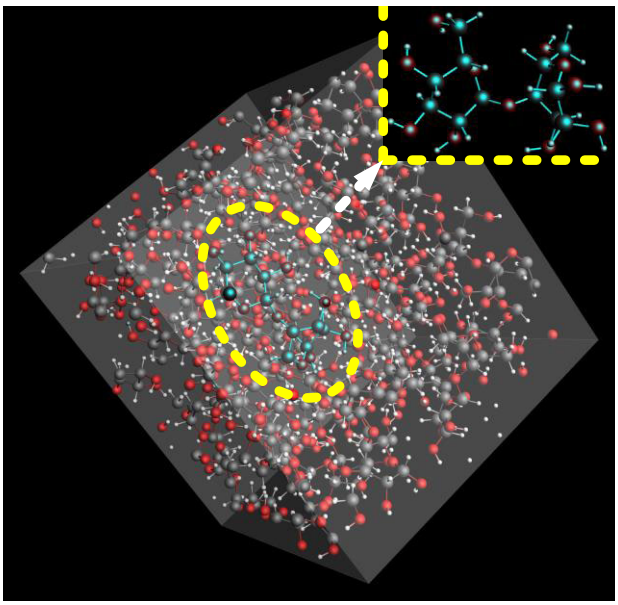

FIGURE 2. Optimized cell model.

in nature) [15], [18]. The treatment process was as follows: the energy of the initial amorphous cell model was minimized under NPT Berendsen ensemble. For the cellobiose studied in this paper had three elements of C, H and O, the CHO.ff force field was selected with a step number of 9000 and a pressure of 500 MP. The optimized size was 22.66 nm × 22.66 nm × 22.66 nm and the density was 1.594 g/cm^3, both of which were approximately equal to the actual density of cellulose in nature [15], [18]. Owing to optimization, the model we constructed was closer to the actual stable model as shown in Figure 2. In the CHO.ff of ReaxFF, the following ten molecular reaction kinetic simulations were carried out: The molecular dynamics simulation of the molecular ReaxFF-MD was carried out using the optimized model to ensure the reliability of the experimental data. Only one variable was changed-the step size, and the other parameters were set exactly the same [28]. Under the NVT Berendsen ensemble, the set temperature was 1900 K, the total reaction time was set to 100 ps, and the number of steps was set to 1000000, 500000, 340000, 250000, 200000, 170000, 150000, 125000, 115000 and 100000, respectively. Since the state of the new atom was obtained by moving the atomic state of the previous moment one step, the range of the set step size should not be too wide. The step size was set to 0.1 fs, 0.2 fs, 0.3 fs, 0.4 fs, 0.5 fs, 0.6 fs, 0.7 fs, 0.8 fs, and 0.9 fs, 1 fs. For there were many uncertain factors in the pyrolysis reaction of transformer insulating paper, Monte Carlo algorithm (Force biased Monte Carlo, MC) was introduced to increase the randomness of the system to ensure that the model can be closer to the real cellulose pyrolysis condition [40], [41]. It was set to start MC once every 500-step molecular simulation. The number of steps per MC operation was 500 steps, and the maximum displacement of atoms between each step of MC is 0.1 fs [31]. Considering the reliability of the simulation results, all calculations were carried out under periodic boundary conditions to ensure that the same breaking environment of each system. The data were recorded every 100 steps in the whole molecular simulation program and used for the analyses of the bonding and breaking process.

IV. RESULTS AND DISCUSSION

A. INFLUENCE OF STEP SIZE ON HEATING

As can be seen from Figure 3, as the step size of point A was 0.1 fs, the system reached a set temperature of 1900 degrees when it ran to about 200 fs. As point B was 0.2 fs, the system reached 1900 degrees when it ran to about 240 fs. As for 0.3 fs at point C, the system needed to run to about 300 fs to reach 1900 degrees. For point D of 0.4 fs, the system ran to about 440 fs to get 1900 degrees. The results showed that the time that the system reached the set temperature increased with the

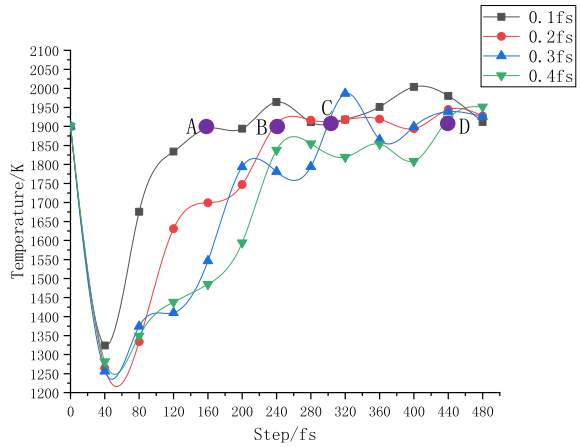

FIGURE 3. Step size and heating rate.

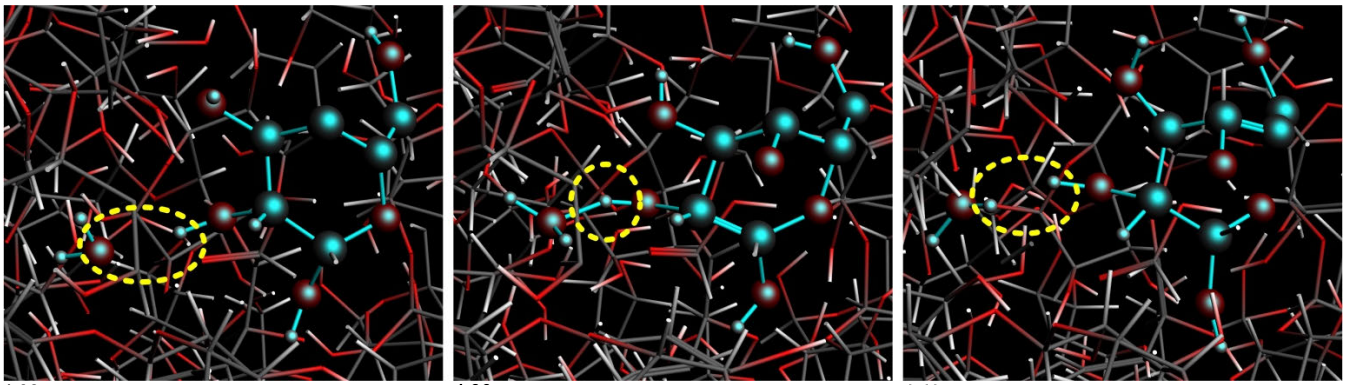

FIGURE 4. Breaking and forming of H_2O at 0.4 fs.

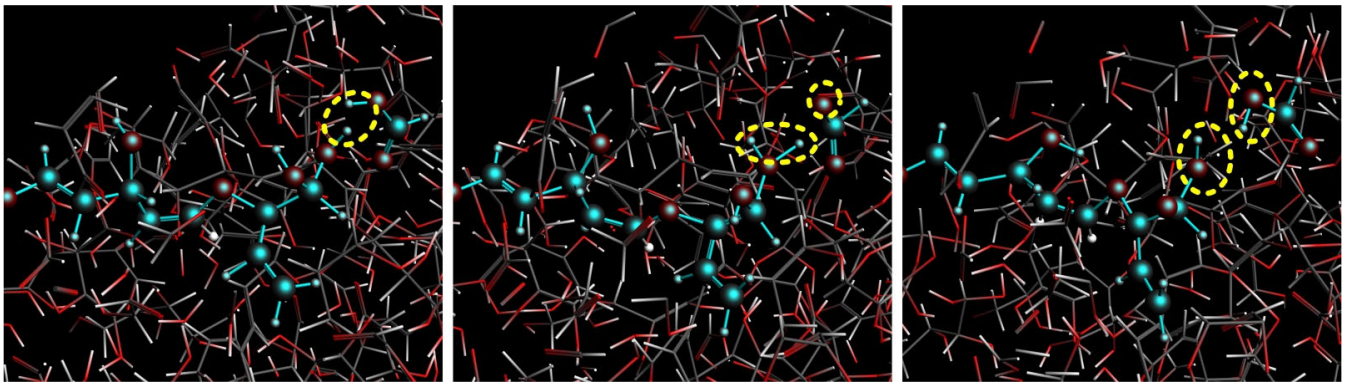

FIGURE 5. Breaking and forming of CH_2O_2 at 0.4 fs.

increase of the step size, and the step size was proportional to the temperature rise to the set degree. Heating the system to the specified temperature in too short a period of time, similar to a pulse, was not conducive to the study on the product. Therefore, it was recommended to increase the step size of the reaction mechanism of cellulose pyrolysis.

B. INFLUENCE OF STEP SIZE ON BOND FORMATION AND BOND BREAKING OF MOLECULES

In order to study the influence of step size on chemical bond formation and breaking, four different step sizes were set under the same conditions [28]. The bonding and breaking of two different molecules H_2O [34], [42] and CH_2O_2 were analyzed, and the conclusions were shown in Table 1. Under NVT Berendsen ensemble, if the set step size was greater than 0.4 fs, the reaction would terminate without a specified number of steps, suggesting that the atomic charge was abnormal in the process of simulation. The electric field was not added

TABLE 1. Bond formation and bond breaking of H_2O and CH_2O_2.

Molecular \ Step	0.1fs	0.2fs	0.3fs	0.4fs	0.5fs-1.0fs
H_2O	yes	yes	yes	yes	——
CH_2O_2	yes	yes	no	no	——

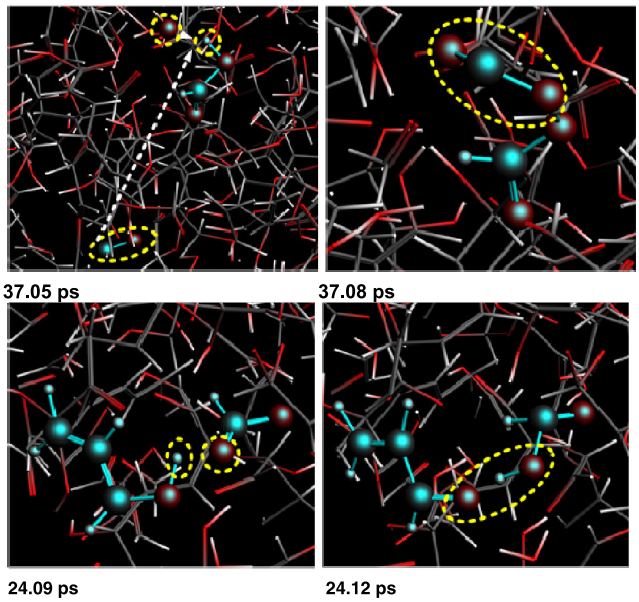

FIGURE 6. Breaking and forming of CH_2O_2 at 0.3 ps.

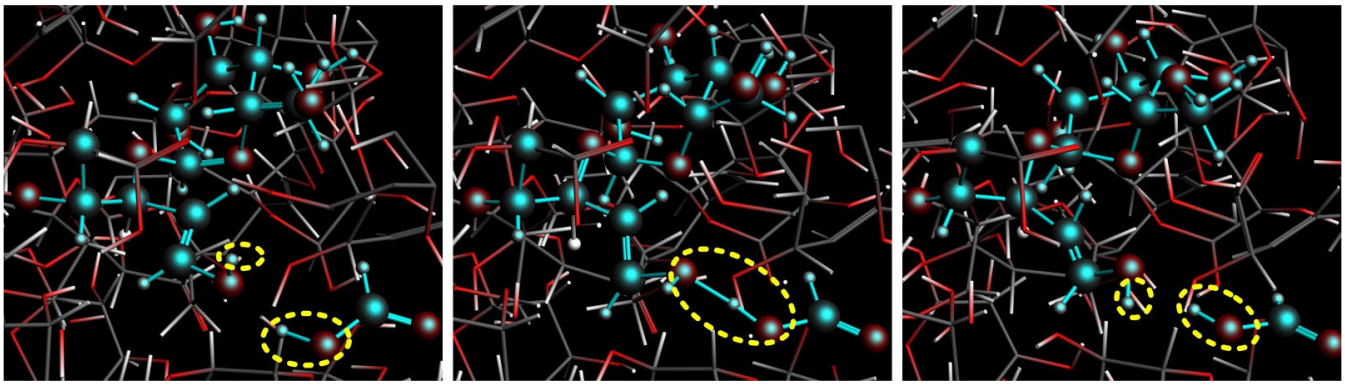

FIGURE 7. Breaking and forming of CH_2O_2 at 0.2 fs.

to the reaction, and the calculation can be restarted from this result. As for this study, once an error occurred, the calculation would be terminated and would not be continued. The cellulose pyrolysis reaction process of 0.5 fs and above was not discussed because the entire reaction process was not completed. For small molecules with simple structure, the chemical bonding and bond breaking can be seen from 0.1 fs to 0.4 fs, while for relatively large molecules such as CH_2O_2, the bonding and breaking process cannot be seen clearly at 0.3 fs and above.

For small molecules such as H_2O with simple chemical bonding, the chemical bonding and breaking of all molecules in the pyrolysis reaction can be seen at 0.1 fs-0.4 fs. Taking the 0.4 fs as an example (Figure 4): the disappearance of water molecules can be observed clearly, and in detail, the oxygen atoms in water molecules combined with one of the hydroxyl atoms on the free radical to form a hydrogen bond. Thus it can be concluded that the bonding breakage of small molecular products can be analyzed by setting the step size to 0.4 fs and below. In the past, the empirical value was generally 0.1 fs, and for small molecules, the step size setting can be increased to reduce the pulse effect.

For larger molecules such as CH_2O_2, when the step size was 0.4 fs, the bonding and breaking process cannot be clearly presented. Only by searching for a changing chemical bond around the molecule can we guess its bonding or breaking mechanism. As shown in Figure 5, at 16.36 ps, there was a single CH_2O_2, while the bond breakage occurred at 16.40 ps, and it was found that the hydroxyl group of an adjacent atomic group had one more hydrogen atom, so it was speculated that the hydroxyl groups on this atomic group took away the hydrogen atom of CH_2O_2. At 16.44 ps, it was observed that the hydrogen atoms in the hydroxyl groups in the adjacent atomic groups disappeared, meanwhile the CH_2O_2 was formed again. Hence, it was judged that the carboxyl group captured one hydrogen atom in the adjacent hydroxyl group to regenerate CH_2O_2.

As shown in Figure 6, when the step size was 0.3 fs, at 37.05 ps, CH_2O_2 disappeared because a combination of oxygen and carbon and oxygen atoms grabbed hydrogen atoms from the hydroxyl group. However, for the step size was too large and the reaction was too fast, it was not easy to see how the bond was broken, so the step size of 0.3 fs was not applicable for macromolecules. At 24.09 ps, an oxygen atom on a group grabbed a hydrogen atom in the hydroxyl group to form CH_2O_2. When the step size was set to 0.2 fs, as shown in Figure 7, the hydrogen atom in the carboxyl group in the CH_2O_2 formed a bond with the oxygen atom in the hydroxyl group in another atomic group to make the CH_2O_2 disappeared, and the newly-formed instable bond broke again to construct another CH_2O_2 molecule.

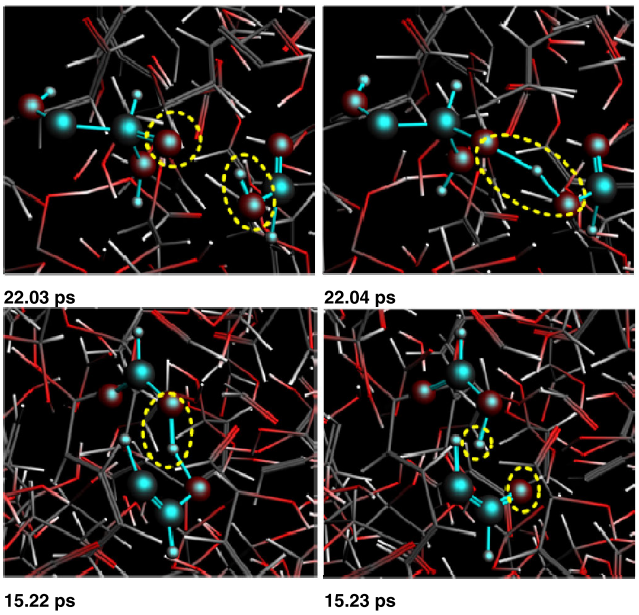

FIGURE 8. Breaking and forming of CH_2O_2 at 0.1 fs.

As shown in Figure 8, the step size was 0.1 fs. At 22.04 ps, the hydrogen atom on the carboxyl group of CH_2O_2 formed a covalent bond with the double bonded oxygen atom on the carboxyl group of the atomic group, so that CH_2O_2 disappeared. At 15.23 ps, the oxygen-hydrogen bond in the radical

was broken to form the molecular CH_2O_2. In summary, when analyzing the bonding or cleavage of the product in the pyrolysis reaction, a step size of 0.2 fs or less was recommended for the macromolecular product, and 0.4 fs or less was for a small molecule product with a simple structure. Appropriate increase of the step size for small molecules cannot only solve the instability of similar pulses when the temperature was heated in a strikingly short time to the specified temperature, but also can reduce the simulation time and improve the research efficiency.

V. CONCLUSION

In this paper, the molecular dynamics simulation of cellobiose pyrolysis at a high temperature, a structural unit in insulating paper, is carried out at the atomic level. The formation and breaking of chemical bonds are analyzed by setting different step sizes at intervals of 0.1 fs, and the relationship between the step size and the heating rate is obtained. The conclusions are as below:

- Under the NVT Berendsen, with the increase of the set step size, the time for the system temperature to rise to the set temperature also increases. At 0.1 fs, it takes 200 fs for the system to heat up to a set temperature of 1900 degrees; at 0.4 fs, the system needs to heat up to a set temperature of 1900 degrees for 440 fs, which is 2.2 times that of 0.1 fs. The results showed that the time for the system to rise to the set temperature is related to the set step size and the step size is proportional to the temperature rise to the set degree.
- The principle of bond formation and breaking of cellulose pyrolysis products is related to the setting step size and the molecular structure of the product. The breaking and bonding of complex molecules such as CH_2O_2 need more steps than simple molecules. In order to study the process mechanism in more detail, it is concluded that the more complex the structure of the product, the step size should be less than 0.2 fs.
- In order to observe the chemical bond formation and breaking of cellulose at 1900 K, the setting value should not be greater than 0.4 fs. Otherwise, the system will report that the atomic charge is abnormal in the simulation process, and the reaction cannot be carried out effectively and correctly.

In this paper, the molecular dynamics simulation of transformer insulating paper cellulose at high temperature is studied from the atomic point of view via the combination of the molecular dynamics simulation and Monte Carlo random calculation method. For small molecules such as H_2O, the larger step size compared with molecules such as CH_2O_2 can be set to observe their bonding and breaking, which can help to reduce the pulse effect under the NVT ensemble, and reduce the simulation time. This study will render a theoretical basis for the subsequent step-by-step setting of the forming and breaking of the pyrolysis reaction product, which simplifies the experimental process.

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