New Insight into Dynamic Mechanical Relaxation in N-butyl-N-(2-nitroxy-ethyl) Nitramine Plasticized Nitrocellulose through Molecular Dynamic Simulations

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New Insight into Dynamic Mechanical Relaxation in N-butyl-N-(2-nitroxy-ethyl) nitramine Plasticized Nitrocellulose through Molecular Dynamic Simulations

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Abstract

We performed dynamic mechanical analysis (DMA) on nitrocellulose (NC) plasticized by an insensitive plasticizer N-butyl-N-(2-nitroxy-ethyl)nitramine (Bu-NENA). NC/Bu-NENA blend shows two mechanical relaxation processes in the temperature ranges of -50~-40°C and 30~40°C, and their variations with deformation frequencies were studied. To explore further the effect of temperature on relaxation, the binary mixture model of NC/Bu-NENA was constructed, and molecular dynamic simulations were conducted. The simulated mean square displacements (MSD) show abrupt increase in the temperature range of -50~-40°C and 30~40°C, which are consistent with those of the two relaxation processes observed in the DMA curves. Moreover, the free volume (V_{free}) and torsion energy obtained from molecular dynamic simulations exhibit distinct increase at the temperature above 30°C and -50°C respectively, reflecting the sudden enhancements on the mobility of polymer chain elements and the rotation of molecular bonds. Furthermore, the radial distribution function (RDF) associated with the intermolecular interactions reveals that the intensities of both hydrogen bond and van-der-vaals forces decrease with the increase of temperature, which is responsible for the decrease of storage modulus at high temperature. These computational and experimental studies reveal guidance to strengthening the NC base propellants in broad temperature range.

Key words

Nitrocellulose, mechanical relaxation, molecular dynamic simulations

Nitrocellulose (NC), an energetic binder has been widely used in the fabrication of double base propellants, wherein NC serving as a major ingredient exerts great influence on the physical properties of propellants. Specifically, the mechanical properties of propellants at varied temperature play important role in dominating their practical applications. Nevertheless, NC features uneven rigidity nature, making it very difficult to uniformly mix with other ingredients, thus imposing adverse effect on the mechanical properties. In general, nitroglycerin (NG) was used as plasticizer to improve the processing of NC, and the influence of NG on the physical properties of NC has been well investigated (Yang et al. 2017; Ma et al. 2008; Qi et al. 2019).

Recent works revealed that the replacement of NG with insensitive energetic plasticizers has significantly reduced the friction and impact sensitivities, which facilitate to improving the process security and the vulnerability of propellants (Chakraborthy et al. 2004; Damse et al. 2009; Yang et al. 2014; Reese et al. 2014) Moreover, the employment of insensitive plasticizers can strengthen the thermal stability and lower the vaporization rate compared with NG plasticizer, which contributes to enhancing mass homogeneity of NC base propellants during storage (Zhao et al. 2017; Wilker et al.2007; Ou et al. 2018). It is well known that the physical properties of NC show strong dependence on the additives used in propellants. In the reported works, the influences of solid fillers and plasticizers have been described for analyzing the physical properties of NC base propellants (Sun et al. 2018; An et al. 2012; Wu et al. 2020; Wang et al. 2019; Qi et al. 2020).

The static stress-strain relationship at different temperatures was depicted, which offer rational explanation for the factors that affect the physical properties. In comparison to static mechanical analysis, the changes in physical properties of NC can be better identified in the dynamic mechanical analysis (DMA), as DMA measurements possess advantages in identifying distinguishing the relaxation of polymer chains associated with the storage modulus (E'), loss modulus (E"), and tan δ (ratio of E"/E')

as a function of temperature (De Paula et al. 2019). The influences of plasticizers on the dynamic mechanical properties of NC base propellants have been investigated, revealing the existence of two principal relaxation processes at high (+40°C) and low (-35°C) temperatures (Warren et al. 1988; Baker et al. 1987). Although, many applications of DMA have been explored for the study of dependence of the mechanical forces and energy loss on the temperature and frequency, fewer attention was paid on the manner whereby the plasticizers impact the transition behavior of NC. Especially, the molecular mechanism of the influence of plasticizer on the relaxation of NC is not well understood. As a main factor affecting the relaxation, the interaction between NC and plasticizer could be evaluated more precisely at high plasticizer content, which will impose great safety issues when using sensitive plasticizers.

In this contribution, we replaced NG with an insensitive plasticizer N-butyl-N-(2-nitroxy-ethyl)nitramine (Bu-NENA) to overcome the safety restriction, and prepared NC/Bu-NENA composite with a mass ratio of 1:1. The dynamic mechanical properties of NC/Bu-NENA were studied as a function of temperature, aimed to distinguish the relaxations and transitions in the plasticized NC. Furthermore, molecular dynamics simulations were conducted to study the role of Bu-NENA in the relaxation behavior of NC, and the simulated parameters such as mean squared displacement, free volume, and potential energy were correlated with the motion of molecular chains and chemical bonds. These experimental and simulative studies enable better explanation of the dependence of physical properties on the temperature, and reveal ~~great~~ potential guidance to strengthening the NC propellants in broad temperature range.

N-butyl-N-(2-nitroxy-ethyl) nitramine (Bu-NENA) was purchased from Haohua Chemical Group, Co., Ltd., China. Nitrocellulose (NC) was purchased from Sichuan Nitrocell Co., Ltd., China. All the reagents were used without further purification unless otherwise specified. The nitrocellulose pellets (nitrogen content 11.9wt%) were mixed with the same mass of Bu-NENA in a vertical kneading machine (HKV-II) under vacuum for 30 min. Then, the mixture was solidified for 96h at 70ºC, and the as-prepared composite was cut into specimen with a dimension of 4mm×4 mm×12mm. The dynamic mechanical analysis (DMA) was carried out on a DMA instrument

(Netzsch 242) in a tension model. The DMA measurements ran at the temperature ranging from -70 ºC to 110ºC with a heating rate of 4K · min^{-1}, and the DMA curves were collected at the deformation frequencies of 1, 2, 5, 10 and 20 Hz, respectively.

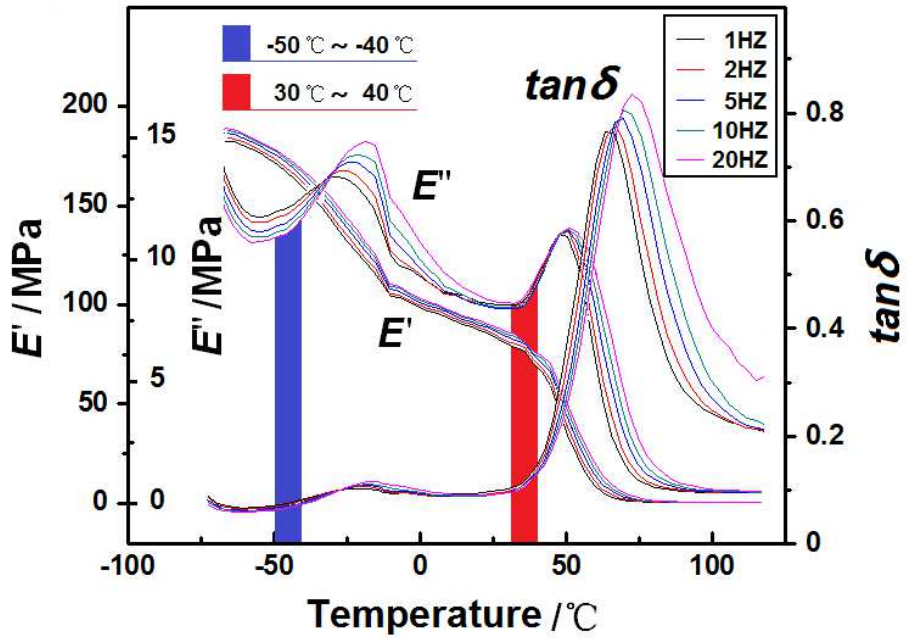

Fig.1 Dynamic mechanical curves of NC/Bu-NENA

Fig.1 shows the storage modulus E', loss modulus E'' and the loss factor tan δ of the NC/Bu-NENA composite obtained by DMA in tension mode at five different deformation frequencies. All the loss factor curves acquired at constant heating rate of 4k · min show two peaks centered at low and high temperature correlating to the β and α transitions respectively. The β transition has been attributed to the motion of plasticizer molecules associated with NC side groups, whereas the designation of the α transition is less definite, which is assumed as the relaxation of the stiff NC chain rather than a transition (Warren et al. 1988; Baker et al.1987). The splitting phenomenon was not observed in the α peak, indicating that unique state exists in the glass transition process of the NC/Bu-NENA specimens.

Dynamic mechanical properties NC/Bu-NENA show strong dependence on the relaxation of polymer chains near the glass to rubber transition temperature (T_g). Typically, increasing temperature above T_g allows to increase the free volume of polymer, and thus lead to increased molecular mobility. Oppositely, the NC/Bu-NENA

composite becomes hard and brittle below T_g, because of the reduced free volume at low temperature. In fact, DMA has been preferentially employed to study the relaxation and transition that are undetectable by DSC and TMA (Zhang et al. 2013; Saba et al. 2018; Meng et al. 2014; Bohn et al. 2015; Seyidoglu et al. 2020).

Herein, DMA was conducted at a constant heating rate and at different deformation frequencies. The storage (elastic) modulus (E'), loss (viscous) modulus (E") and loss factor (tanδ) equivalent to the ratio E"/E' were recorded as a function of temperature. As shown in Fig.1, the storage modulus (E') gradually decreases with the increase of temperature in the case of all frequencies, implying that the energy storage per unit strain was lowered at high temperature. In general, mechanical relaxation occurs in the glass transition process of polymers, during which the flexibility and mobility of molecular chains undergo prominently changes. The decreased storage modulus is an indication of the increased mobility of NC/Bu-NENA chains at high temperature. Oppositely, increasing the deformation frequency in DMA leads to an increase of the *E*' value at the same temperature, and this is due to the strain-rate hardening effect of polymer chains at high deformation frequencies (Bohn et al. 2015;). The curves of loss modulus versus temperature show two peaks centered at around -25°C and 50°C, which are associated with the maximum energy loss arising from the internal friction and molecular rearrangement processes of polymer chains.

Moreover, the loss modulus curves show two inflection points at the temperature of -50°C and 30°C, corresponding to the onset temperatures of multiple relaxations of polymer chains. At the temperature of -50 °C, the side chain elements of NC start moving as the imported thermal energy overcomes their energy barrier. Increasing the temperature up to 30°C initiates the cooperative motion of numerous chain segments, which allows large scale deformation of NC/Bu-NENA, and hence lowers the storge modulus. Besides, the tanδ curve representing ratio of E"/E' was plotted against temperature, and the maximum value of tanδ is a measure of glass transition temperature. The tanδ curves show two peaks reflecting the damping characteristics of NC/Bu-NENA at low and high temperatures, respectively. In fact, the intrinsic properties of polymer binders or plasticizers experience substantial change from glass

state to rubber state by increasing temperature, which is an indicator of the change in the molecular mobility of the polymeric networks during the glass-rubber transition. Moreover, the maximum tanδ value and its corresponding glass transition temperature increase with the increase of frequency, which is due to the freeze of the polymer chain elements at high tensile frequency.

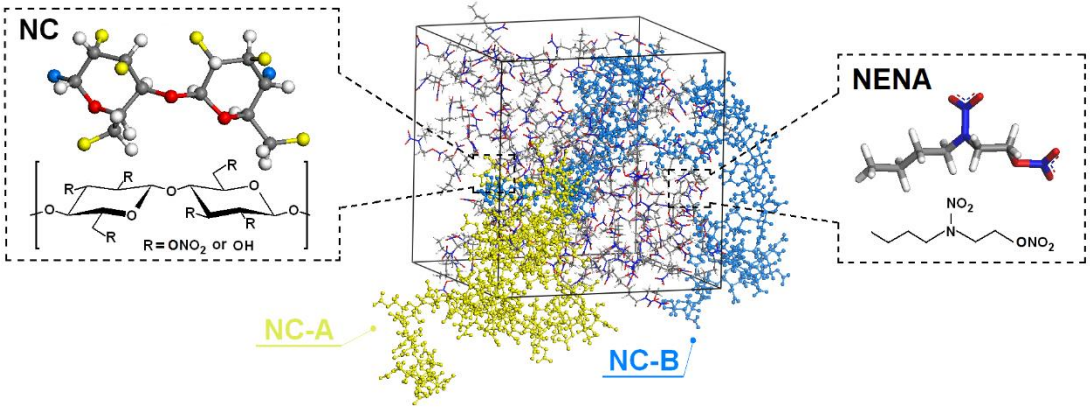

Fig.2 Molecular structures of NC and Bu-NENA, and the simulated primitive cell of the NC/Bu-NENA blend, NC-A and NC-B represent two different molecular chains of NC in the constructed model.

To gain deep insights into the variation of dynamic mechanical properties from the perspective of the motion of polymer chains, and explain the effect of temperature on the mechanical relaxation process of NC/Bu-NENA at molecular level, the molecular dynamic simulations were performed to study the atomistic model of NC/Bu-NENA by means of Material Studio (Accelrys) according to our previous work (Qi et al. 2019). Given that the nitrogen content of NC is 11.9wt%, the average degree of substitution of NC is calculated to be 2.2 according to the reported equation (Jamal et al. 2020)

$$DS = \frac{1.62 \times \%N}{14 - 0.45 \times \%N}$$

Regarding the NC/Bu-NENA blends, the molecular chains of NC are coarse grained into the model of Gaussian chains, which are constituted by numerous basic NC units. Herein, 320 and 240 basic NC units were taken in the NC-A and NC-B Gaussian chains respectively. The simulations were conducted under a COMPASS (Condensed-phase Optimized Molecular Potentials for Atomistic Simulation Studies) force field. In detail, 2 molecular chains of NC were mixed with 98 molecules of Bu-NENA in the simulated

primitive cell as shown in Fig.2, given that the mass ratio of NC and Bu-NENA is 1:1. The NC/Bu-NENA was optimized by a 5000 step energy minimization to regulate unbalanced configurations. To achieve an equilibrium structure, a 100ps molecular dynamic simulation was performed on the system in the NPT ensemble under constant pressure with a time step of 1fs. The simulations were carried out at different temperatures of -80, -70, -60, -50, -40, -30, -20, -10, 0, 10, 20, 30, 40, 50, 60, 70 and 80°C, and the simulation results including the mean-square displacement, volume distribution, potential energy and radial distribution function were obtained.

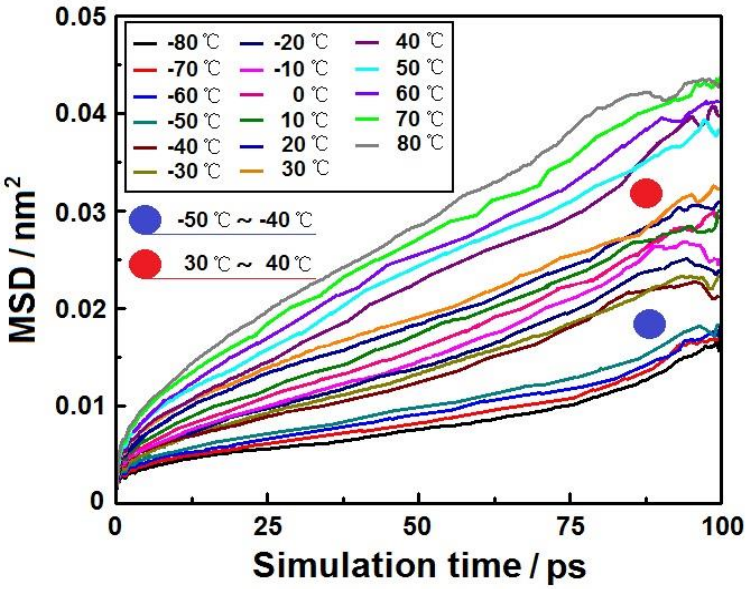

Fig.3 Mean-squared displacements of NC/Bu-NENA.

To probe the dynamics of Bu-NENA molecules and assess its influences on the relaxation of NC, the mean-squared displacements (MSD) of plasticizer was estimated through molecular dynamics simulations. MSD is a statistical average of the trajectory of plasticizer molecules with simulation time and can be estimated from the production stage of MD simulation, during which the drift of the center of mass of the simulation box is subtracted, and expressed by the following equation:

$$MSD=\left\langle \left| r_t - r_0 \right|^2 \right\rangle$$

where r_0 and r_t represent the positions of the centre of mass of plasticizer molecules at time *0* and *t* respectively, and the angular bracket represents the ensemble average. Typically, MSD allows to describe the diffusion of plasticizer in NC/Bu-NENA blend.

In brief, when the blend is frozen then MSD saturate, and the kinetic energy is too low to achieve thermal diffusion. Nevertheless, if the blend is not frozen then the MSD increases almost linearly with time, which allows to understand the dynamics of the blend from the slope of the MSD.

Herein, molecular dynamic simulations were conducted to determine the MSD curves versus simulation time at -80, -70, -60, -50, -40, -30, -20, -10, 0, 10, 20, 30, 40, 50, 60, 70 and 80 ^{o}C respectively, as shown in Fig.3. Notably, the simulations at higher temperature result in larger MSD values. The MSD curves show approximately linear relationship with respect to the simulation time, and the slop reflecting the mobility of plasticizer molecules increases with the increase of temperature. More interestingly, the slops of MSD exhibit two distinct increases in the temperature ranges from -50^{o}C~-40^{o}C and 30^{o}C~40^{o}C, suggesting that the molecular mobility of the plasticizer was significantly enhanced, which may be due to the sudden increased free volume of NC polymer in those two temperature ranges. The increased free volume facilitates to promote the relaxation of NC polymer chains. The relaxation processes disclosed in the MSD simulation are consistent with those revealed in DMA curves, verifying that NC/Bu-NENA possesses two principle mechanical relaxations in the temperature between -80 and 80^{o}C. It should be noted that the temperature at which the mechanical relaxation happens revealed in MSD is about 25^{o}C, which is lower than the glass transition temperature determined from the maximum tanδ value, and this delay is due to the viscoelastic nature of NC/Bu-NENA that leads to strain hysteresis of polymer chains under the action of external force.

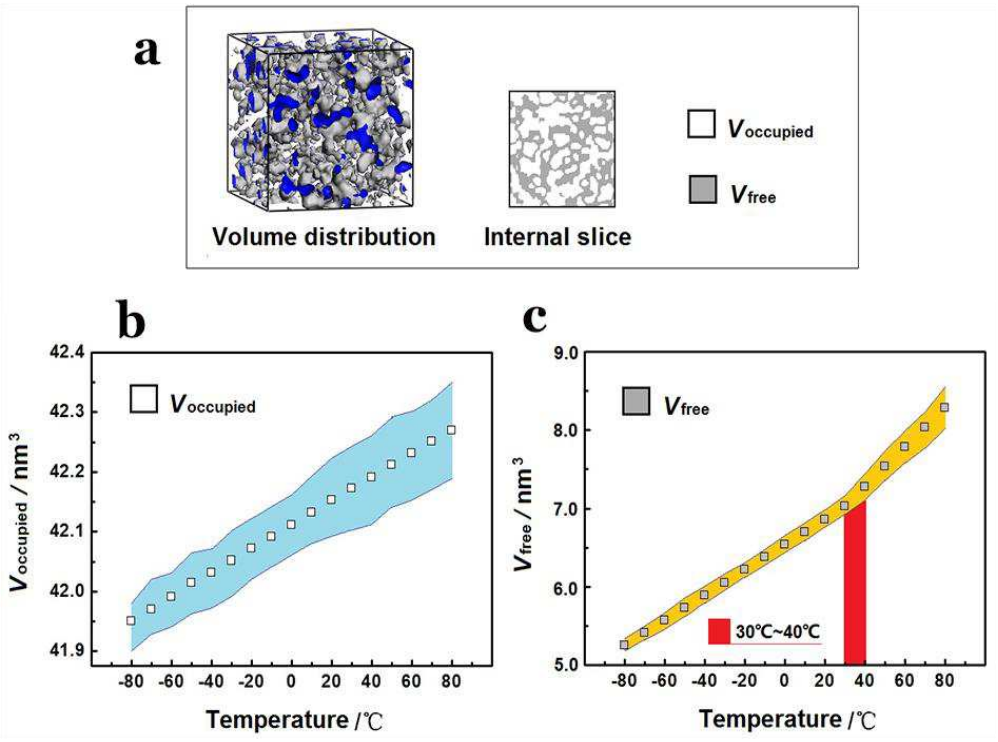

Fig.4 Volume distribution of NC/Bu-NENA at different temperatures

It is well established that the total volume of polymer consists of the volume occupied by polymer chains ($V_{occupied}$) and the interstitial spaces among the chains, namely free volume (V_{free}), among which V_{free} strongly affects the motion and relaxation of molecular chains. Hence, to further correlate the dynamic mechanical properties with the motion of polymer chains, the molecular dynamic simulations were conducted to study the volume distribution in the NC/Bu-NENA blend. Fig.4a shows the volume distribution of NC/Bu-NENA at different temperature, where the distribution of $V_{occupied}$ and V_{free} are visualized as white and grey areas in the cubic box, and the blue area indicates the cross section of free volume.

Fig.4b and c show the variation of $V_{occupied}$ and V_{free} with respect to temperature. The V_{free} value exhibits substantial increase from 5.258 to 8.287 nm^3 with the temperature increasing from -80 to 80°C, whereas the $V_{occupied}$ value shows only slightly increase from 41.951 to 42.270 nm^3, attributed to the unveiling of the overlapped volume at high temperature. It is noteworthy that the V_{free} value shows a faster increase when the temperatures are higher than 30°C. This result indicates that a critical temperature exists, at which the mobility of NC molecular chains significantly changes, resulting in an acceleration of the mechanical relaxation of the molecular chains of NC. This variation

of free volume is consistent with the simulation for the mean-squared displacements (MSD) in the temperature range of 30~40°C, which allows to clarifying the mechanical relaxation and transition processes occurred at high temperature revealed in DMA. Actually, the sudden change of free volume is closely related to the glass transition of NC/Bu-NENA blend. The fast increase of free volume enables cooperative motion of numerous chain segments, and allows large scale deformation, with consequence of significant reduction of modulus.

Potential energy

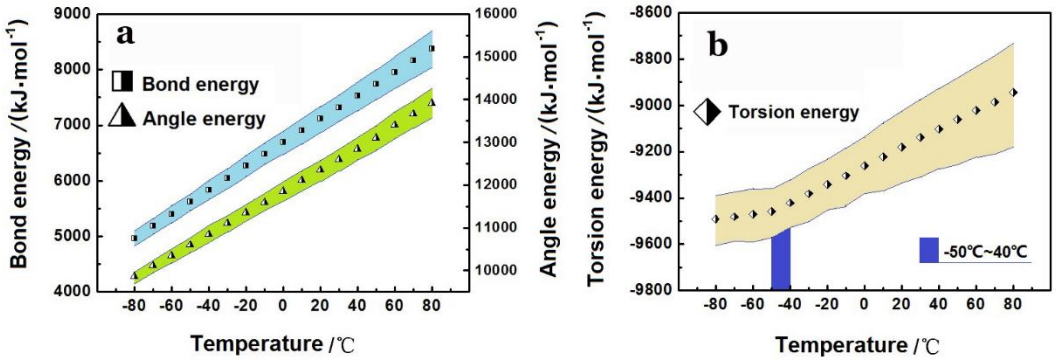

Fig.5 Potential energies of NC/Bu-NENA at different temperatures

In addition to free volume and mean square displacements, the potential energies consisting of intramolecular and intermolecular energies acquired from molecular dynamics simulation was adopted to study the mobility of polymer chains. Considering the arrangement of atoms in the molecular structure of NC/Bu-NENA, the potential energy consists mainly of bond energy, angle energy and torsion energy of the molecular chains. Fig.5 shows the potential energy of NC/Bu-NENA as a function of temperature. Obviously, all the potential energies increase with the increase of temperature, suggesting that the intermolecular interaction between NC and Bu-NENA was enhanced. The bond energy and angle energy exhibit almost linear increase as shown in Fig.5a, whereas the torsion energy displays two steps increase separated by the temperature region from -50 to -40°C, as highlighted in Fig.5b. This phenomenon shows a good agreement with the simulated mean square displacements at low temperature, indicating that the torsion movement in the molecular chains happened due to their shortened relaxation time in the temperature ranging from -50 to -40°C. This result can also explain the mechanical relaxation process taking place at the low

temperature revealed in DMA.

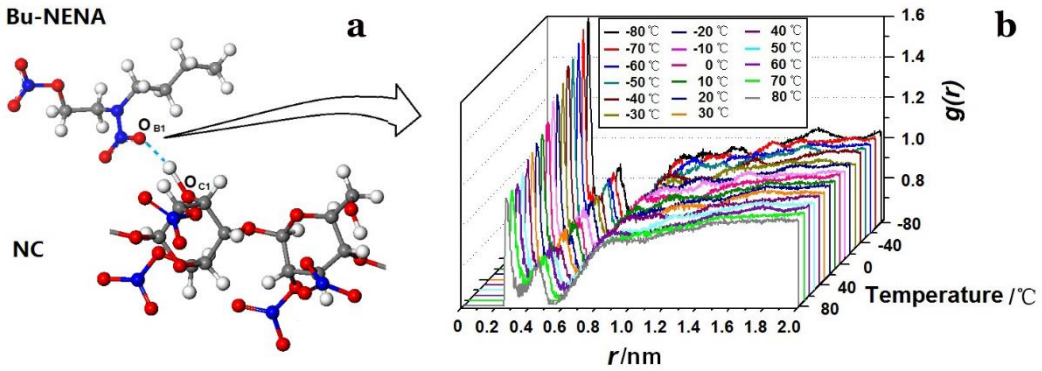

Fig.6 RDF curves for O_{C1}-O_{B1} atom pair in the NC/Bu-NENA at different temperatures.

To understand the effects of temperature on the dynamic mechanical properties at the molecular level, the pair correlation function (PCF) was used to describe the intermolecular interaction between NC and Bu-NENA. PCF allows to depict the local spatial correlation of atom pairs, and enables measuring the atom density at certain distance from a reference atom. The pair correlation function can be defined as

$$g^{(2)}\left(r,r'\right)=\left\langle v^{(2)}(r,r')\right\rangle \Big/ n^2 \tag{263}$$

where n is the particle number density, and $v^{(2)}(r, r')$ is pair density, which is expressed as:

$$v^{(2)}(r,r')=\sum_i\sum_j \delta\left(r_i-r\right)\delta\left(r_j-r'\right) \quad (i\neq j) \tag{266}$$

in which r_i and r_j is the position vector of atoms i and j respectively. The types and intensities of interaction force can be judged from the peak position and peak height in the PCF curves. Generally, the r values of hydrogen bond and van der waal force are in the range of 0.20-0.31 nm and 0.31-0.50 nm, respectively.

Herein, we studied on the potential interaction forces formed between the hydroxyl groups of NC and the nitrate groups of Bu-NENA. For simplicity, the oxygen atom in the hydroxyl group of NC was labeled as O_{C1}, while the oxygen atom in the nitro group of Bu-NENA was denoted as O_{B1}. Fig.6a illustrates the interaction between NC and Bu-NENA at molecular level, and the variations of O_{C1}-O_{B1} atomic force with respect to

temperature are shown Fig.6b. The RDF curve for the atom pair O_{C1}-O_{B1} at -80ºC displays a peak at 0.259nm, corresponding to a g(r) value of 1.606, which indicates that the type of atomic force O_{C1}-O_{B1} belongs to the hydrogen bond. The second peak observed at r=0.447nm corresponds to the g(r) value of 0.887, and this peak is associated with the van der waal forces between the oxygen functionalities in NC and Bu-NENA. Notably, the intensities of both peaks in RDF curve decrease with the increase of temperature, suggesting that both hydrogen bond and van der waal force decrease. The decrease of intermolecular interaction enables enhancement on the flexibility of NC molecular chains, which interprets the decrease of storage modulus with increasing temperature.

In summary, NC/Bu-NENA was prepared through a conventional kneading approach. DMA reveals two main relaxation processes at high and low temperatures, which correlate with the movements of the molecular chains and the torsion of the molecular bonds. Molecular dynamics simulations were conducted to verify the experimental results. The simulated mean square displacements and free volume demonstrate abrupt increase in the temperature range of 30~40ºC, which confirm the mechanical relaxation and transition processes revealed in DMA. Moreover, the simulated torsion energy displays significant increase in the temperature range from -50 to -40ºC, which verifies the torsion relaxation of molecular bonds revealed at the low temperature range of DMA curves. Besides, the lower intermolecular interaction at higher temperature observed in radial distribution function is the reason for the decreased storage modulus of NC/Bu-NENA with increasing temperature.

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Conflict of interest The authors declare that they have no conflict of interest.

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