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Study on failure mechanism of Cu-polyethylene-Cu sandwich structure by molecular dynamics simulation

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ABSTRACT

The tensile failure mechanism of Cu-Polyethylene (PE)-Cu (CPC) sandwich structure was clarified by molecular dynamics (MD) simulations subjected to a uniaxial tensile loading at microscopic scale. The sensitivity analysis of parameters such as mixing rules in describing the interaction between the wall (Cu) and the sandwich layer (PE), model size, relaxation time for equilibrium and initial velocity distribution was carried out to verify the rationality of modeling. The evolutions of stress-strain relationship and each potential energy component were provided to describe the failure process of the structure. The peak of non-bond energy shows a delay compared to the yield point in stress-strain curve, which coincides with the local maximum point of the trans-fraction curve of dihedral angles. After that, an inflexion appeared in the trans-fraction curve indicates an energy transport process, which corresponds with the slope change of the stress-strain curve. It is assumed that the dihedral distribution plays a crucial role in the damage process of CPC structure. In addition, the temperature field and the density profile were adopted to predict the position of damage initiation, which was confirmed by the microstructure evolution. The intrinsic thickness-dependence of CPC was explored by taking the coupling effect of bridging and entanglement into account, which is in reverse proportion with the yield strength of CPC.

1. Introduction

Interfaces between polymers and metals are common in industrial systems, such as in adhesive structures for automotive and aircraft applications, microelectronic device packaging and coatings [1]. Generally, an interface forms with adhesion when the physical and chemical reactions occur between two phases (matrix and polymer layers). As an effective component to transfer stress and bear the structural loadings, the bonding structure composed of adhesive and metal matrix is the focus of our research.

The two-phase interface structure formed by the combination of metal matrix and polymer, as a weak link, is most likely to occur damage during the whole structure bearing a large loading. The failure process of the two-phase structure is often different from that of a single phase, which depends not only on the physical properties such as toughness, strength and stiffness of the polymer, but also on the nature of entire structure [2]. In order to make the bonding structure more economical and reliable, it is necessary to deeply understand and clearly describe its mechanical behavior under loading, so as to provide a theoretical basis for optimizing the design related to the bearing capacity of the component.

The general polymer materials have both the characteristics of entanglement and cross-linking [3]. The micro-holes in the polymer layer do not aggregate and generate large cracks when the damage occurs. In contrast, fibrillation occurs due to a certain mechanism in failure process, which is described as macroscopic *silver craze* [4]. In addition, metal substrates are typically several orders of magnitude stronger than polymers. Three main categories as cohesive failure (failure in the polymer layer), adhesive failure (debonding at the polymer-adherend interface) and mixed failure (combined with cohesive-adhesive failure) are taken into account, irrespective of the failure of the metal matrix [5].

In the early studies, research scholars tended to accumulate and analyze experimental data and tried to establish analytical or numerical models. The load-testing modes for the adhesive system mainly include tensile, shear and split. Accordingly, the relationship between the local separation displacement and the traction stress at the bonding interface can directly express its mechanical response. The experimental data can be integrated into a specific mathematical model to obtain the normal or shear separation strength [6,7]. Liao et al. [8] carried out the

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experimental and finite element (FE) calculation for single-lap adhesive joints, which presented the different rupture initiation positions under impact and static tensile loadings. As one of the important parameters during the bonding process, the thickness of the adhesive layer, especially with a small value, has a great influence on the interfacial strength. When the thickness exceeds a certain critical value [9], the area near the wall will not be affected by plastic deformation [10]. In addition, the design and fabrication of fiber-reinforced composite structures [11–13] can also give us the inspiration about the design of adhesive joints for improving their properties under mixed loadings. As for the analytic models, the descriptions of the two-dimensional stress distribution in interfacial structures at the continuum level are favored largely. Most of existing analytical models focus on linear elastic solutions of polymers and matrix under different external loadings [14,15]. The difficulty for obtaining the analytic solution increases with the uprising complication of the forms used to complete the analytic model. Therefore, researchers usually simplify actual problems as the idealized ones to fit for these developed analytical solutions. In the aspect of numerical models, the cohesive zone model (CZM) can characterize the elastic-plastic behavior of the adhesive structure [16] to predict the overall failure of adhesive bond, which can be easily implemented by FE calculation. Liao et al. [17] studied the load-bearing capacity and the damage level of the double scarf joint (DSJ) by FE method using the mixed-mode CZM with a bilinear shape. However, the cohesive parameters for the same loading method of different materials or the different loading methods of the same material are very different. Correspondingly, many researchers contributed to study the influence of cohesive parameters on the prediction accuracy of the overall mechanics behavior of the adhesive structures [18–20].

Considerable results were obtained aiming to the behaviors of interfacial structures at the macro scale. However, whether in experimental or theoretical studies, the existing characterizations of interface damage mainly focused on macroscopic continuum mechanics and phenomenological hypothesis. For lacking of detailed description of the objective physical image, it is necessary to seek the intrinsic mechanism of the phenomenon of the whole damage process.

Molecular dynamics (MD) simulation can be chosen to study the failure behavior of interfacial structures at atomic scale. Alder and Wainwright [21] adopted the hard-sphere model to study the equation of state for gases and liquids. As development of the basic theories such as computing method and force field, MD method has been gradually applied to the fields of structure and engineering. Some investigators discussed the relationship between structural failure and temperature, interfacial interaction, loading rate by MD [22], whose results agree well with experimental data [4]. Therefore, MD method is an ideal solution to capture the mechanical behavior of the adhesive structures with extreme small thicknesses. In addition, this method can also be adopted to simulate the response of the structures in extreme environments like impact loadings, thermal circumstances, etc. It provides the possibilities of clearly understanding the behavior of polymers at microscopic scale, and grasping the relationship between the chemical structure and physical properties of polymers. Fan et al. [23] obtained the traction-separation relationship of the epoxy/copper interfacial structure using MD method. The constitutive relation was applied to FE analysis in CZM model. Zhou et al. [24] employed this strategy to calculate the cohesive parameters of brittle materials in mixed mode. These studies provide an idea to examine the multi-scale coupling of polymer/metal interface.

The failure mode of an interfacial system containing organic and inorganic materials usually depends on several aspects [9], such as the properties of the material itself (lattice parameters, glass transition temperature, etc.), the constraint effect (adhesive size, interfacial adhesion area, the degree of bonding uniformity, etc.) as well as the external conditions (loading rates, temperature, etc.). The failure of an interfacial system is essentially the result of multi-factor coupling. In previous studies, we analyzed the coupling effect of adhesive type and geometry (adhesive thickness and scarf angle) on the mixed-mode failure of DSJ using dimensional analysis and FE method (CZM) [25,26]. However, the intrinsic mechanism of these influential parameters is still unclear owing to the limitation of the scale. Explorations in smaller scale like atomic- or molecular-scale should be carried out to provide multiscale understandings. There were several investigations on the mechanical responses of a sandwich structure (adhesive-like) consisting of amorphous polyethylene (PE) chains and face-centered cubic crystal copper layers (Cu-PE-Cu, CPC) in the framework of MD, and the branching and cross-linking of the polymer layer were out of consideration for simplicity. However, there were still some problems. The overall sizes in simulations are too small to represent the characteristics of real systems [5]. The description of the relationship between PE interface size and overall strength is too brief [9] to reach a quantitative understanding on the failure mechanism. Moreover, an intuitive description of the failure process of the interfacial system is eagerly to be developed.

The paper is organized as follows. In Section 2, the modeling procedures of bulk amorphous PE and CPC sandwich are described. In Section 3, the damage evolution until failure of CPC sandwich is simulated. The corresponding methods of characterization are also provided. More importantly, the thickness effects coupling with entanglement and bridging are discussed.

2. Simulation method

2.1. Force field

Taking efficiency and accuracy into account as much as possible, a -CH_2- monomer (and the terminal CH_3- monomer) in a PE chain was treated as a *united atom* (UA) [27,28]. Copper layers were considered as rigid bodies without interactions between inner atoms. Accordingly, the atoms in copper layer move as a single group. At the interface of two materials, we assume that there is only the non-bond interaction between copper atoms and PE UA. All MD simulations were implemented by LAMMPS coding [29], and all the visualization operations were performed by OVITO [30].

Dreiding potential [31] was selected to characterize the intra- and inter-molecular interactions of the PE chains, which are with four terms, namely bond stretching (with a subscript of bond), bond angle changing (with a subscript of angle), dihedral angle rotation (with a subscript of dihedral) and intermolecular interactions (with a subscript of non-bond). The total potential energy (with a subscript of total) and each component are expressed by Eqs. (1)–(5), respectively.

$$E_{\text{total}} = E_{\text{bond}}(r) + E_{\text{angle}}(\theta) + E_{\text{dihedral}}(\phi) + E_{\text{non-bond}}(r) \tag{1}$$

$$E_{\text{bond}} = \frac{1}{2}K_b(r{-}r_0)^2 \tag{2}$$

$$E_{\text{angle}} = \frac{1}{2}K_\theta(\theta{-}\theta_0)^2 \tag{3}$$

$$E_{\text{dihedral}}(\phi) = \sum_{i=0}^{3} C_i(\cos\phi)^i \tag{4}$$

$$E_{\text{non-bond}}(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right], \quad r \leqslant r_c \tag{5}$$

where K_b and K_θ are the stiffness constants of the bond length and the bond angle, respectively. r_0 and θ_0 are the equilibrium bond length and the bond angle, respectively. C_i (i = 1, 2, 3) represents the coefficients in Eq. (4) to describe the dihedral angle.

Intermolecular potential energy, as shown in Eq. (5), describes the interactions among the PE chains themselves and that between the copper atoms and the PE UA. Expressed in terms of Lennard-Jones (LJ) 12-6, σ in Eq. (5) defines the equilibrium distance between atoms as the energy is minimum for $r = 2^{1/6}\sigma$, and ε represents the depth of the