**Dear Editors and Reviewers:**

Thank you for your letter and for the reviewers’ comments concerning our manuscript entitled “**Progressive Molecular Rearrangement and Heat Generation of Amorphous Polyethene Under Sliding Friction: Insight from the United-Atom Molecular Dynamics Simulations**” (ID: **la-2020-019497**). Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction which we hope meet with approval. Revised portion are marked in yellow color in the paper. The main corrections in the paper and the responds to the reviewer’s comments are as flowing:

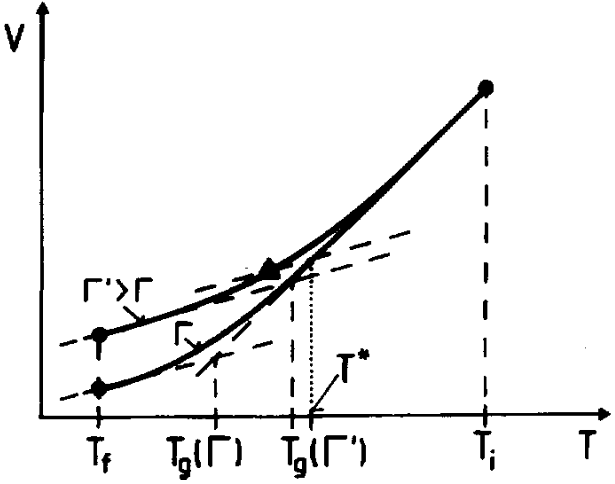
**Reviewer #1**

**General Comment:** In this work, the authors performed coarse-grained molecular dynamics (MD) simulations to study the interfacial friction heating mechanism of amorphous polyethene (PE) in both the Single Sliding Friction (SSF) and Reciprocating Sliding Friction (RSF) modes. They found that the RSF process led to a better heat generation performance that SSF even though there was no apparent plastic deformation. This is different from the traditional view that the plastic deformation is the primary source of heat generation. They attributed the interfacial friction heating enhancement in RSF to the concentrated high-frequency chain motion related to molecular rearrangement.

1. **Comment:** For PE, the yield stress is about 20-30 MPa, which is much high than the loading pressure, which is about 0.2, 0.4 and 0.8 MPa. The authors need to explain what they mean plastic deformation here.
2. **Reply:** Thank you for your suggestion. I will explain this issue in the following two points. Firstly, it is about the setting value of loading pressure. Actually, the loading pressure of 0.2, 0.4 and 0.8 MPa is the pressure setting in LAMMPS MD package, representing the added force on every atom of the group, every bead in the rigid layer in our paper. If these pressures correspond to the pressure of the entire rigid body layer, it must be multiplied by the number of beads in the rigid body layer. For example, the pressure of , , . For macro cognition, it is also a big number, which is also inevitable for MD to study the micro environment. The real area of contact difference between micro/nano scale and macro scale leads to the result that the mean contact pressure in MD can be as large as 3 orders of magnitude larger than the normal applied pressure 1,2. In the past similar friction process under pressure loading using MD method, the pressure value is almost the order of GPa. So in terms of parameter setting, my pressure value setting has no problem. Secondly, it is about the relation between loading pressure value and yield stress of PE or plastic deformation. In our paper, the setting of the pressure value in our paper is actually not directly related to the yield stress or plastic deformation of PE. The loading pressure is used to equilibrium the system before friction simulation, or the friction cannot continue without that, just like in the real situation, we should press the eraser to rub on the paper. The initial state of friction simulation of the system is obtained through pressure balance. Generally speaking, the plastic deformation of polymers needs to be measured in detail through actual tensile experiments, but in the past MD simulations 3–5 have qualitatively analyzed the potential energy curve, bond angle changes and accompanying elastic, plastic deformation. Based on these results, we have carried out a qualitative analysis of the plastic deformation during the friction process and find out that molecular rearrangements play a significant role in heat generation, which is not clearly related to the deformation degree. **Related edits have been added in the manuscript:**

“and normal loading pressure *p* (0.2, 0.4 and 0.8 MPa) was applied to every bead in the rigid body, if transferred to the whole rigid layer, it will be the pressure of *n* (number of beads of the group) *p* (, , .). Due to the real area of contact difference between micro/nano scale and macro scale, the mean contact pressure in MD can be as large as 3 orders of magnitude larger than the normal applied pressure.” (**Loading and Friction Equilibrium section, Paragraph 1, line 5**)

1. **Comment:** For real PE, the melting temperature is 388-408 K and the glass transition temperature is 195 K. But the predicted glass transition temperature here is 300 K. Do the authors really model PE?
2. **Reply:** We totally understand the reviewer’s concern. Firstly, we confirm that our simulation model is polyethylene (PE). Dreiding force field is used in our research which has been wildely used by previous researchers when building PE, and the specific parameters have been given in the table. The glass transition temperature of about 300 K obtained in our paper is consistent with the previous MD simulation results. As for why there is such a difference, it is due to the huge difference between the macro-scale and the molecular scale. Because in molecular dynamics, considering the huge cost of calculation system and operation time, the temperature changing rate of quenching and annealing is much higher than the traditional experimental value, and the higher cooling rate directly leads to the glass transition temperature calculated by MD is dozens of Kelvin higher than the experimental value such as 250 K. A simple schematic of temperature versus system volume at two different cooling rates in Buchholz et al.’s paper 6 was shown below in **Figure 1**, where a higher glass transition temperature . Even so, this does not affect the qualitative analysis of the MD method in a micro environment.



**Figure 1.** Simple schematic of temperature versus system volume at two different cooling rates 6. and represent the calculated glass transition temperature at a higher and cooling rate respectively.

**Related edits have been added in the manuscript:**

“Calculated glass transition temperature is generally higher than experimental result, which is due to the higher cooling rate in micro/nano scale.” (**Glass Transition Temperature section, Paragraph 1, line 14**).

1. **Comment:** The authors discussed chain diffusion and entanglements of PE. Can the authors specify what kinds of PE they simulated, for example chain length, degree of entanglement and gyration radius?
2. **Reply:** The model built in our research is the amorphous PE, Specifically, low-density PE. However, the simulation result is slightly lower than the experimental value for low density PE (), where was obtained, we deem this acceptable for the united atom method potential employed, which previous works 3,5 have also claimed. Chain length in our research is 50. The degree of entanglement is represented by entanglement parameter in our research. The geometric technique of Yashiro et al. 7 was used, which has been widely used in previous MD simulations 3,5 and was adopted to evaluate the inter-chain entanglement. This technique first creates two vectors which emanate from each atom to neighboring atoms that are separated by 10 atoms on the same chain, i.e., one vector that connects atom (*i*) with atom (*i*-10) and one vector that connects atom (*i*) to atom (*i*+10). The angle between these two vectors is calculated for each applicable atom and a threshold parameter of is used to specify if the atom is classified as entangled or not. The atoms classified as entangled via this technique are considered to be constrained by neighboring chains. The averaged chain radius of gyration at equilibrium were equal to about **Related edits have been added in the manuscript:**

“Chain length in our research is 50.” (**Interaction Potentials and Modeling Systems section, Paragraph 2, line 3**)

“In addition, higher temperature results in a lower density of the PE bulk system in the relaxation process, for example, at 100 K and at 300 K, which is close to the Bao, Hossain et al.’sd result. However, the simulation result slightly lower than the experimental value for low density PE (), which contains a high degree of short and long chain, and on the another hand, we deem this acceptable for the united atom method potential employed.” (**Glass Transition Temperature section, Paragraph 1, line 15**)

The averaged chain radius of gyration at equilibrium were equal to about . (**Glass Transition Temperature section, Paragraph 1, the last line**)

1. **Comment:** The simulation cell size is too small. The authors need to assess/examine the size effects.
2. **Reply:** It is really true as you commented that the simulation cell size is too small if compared with the experiment environment, but such a small simulation cell size is not alone in similar friction simulation using MD method.For example, the cell size is in Dai et al.’s multi-layer model 8, simulation box size with in Dai et al.’s sandwiched model 9. The reason why we adopted the size of in this paper is that on the one hand, we refer to the previous friction research, and on the other hand, it is indeed limited by our own computing resources. In our paper, we are currently only able to simulate a small system, even so, we can get similar results in a good agreement with previous studies. If the degree of polymerization and the number of polymer chains are increased, it also means that the calculation cost will increase, but this is also worth paying attention to. Thanks you for your suggestion, we will consider the impact of the degree of aggregation after the subsequent computing resources are satisfied.
3. **Comment:** In calculating the temperature (Eq. (3)-(8)), the authors used the number of atoms. Do they mean the number of beads? How is the calculated temperature converted to the real physical temperature?
4. **Reply:** Thank you for your rigorous consideration, and we should apologize for some of our vague writing, which may lead to inconvenience to readers' understanding. When calculating the temperature (Eq. (3)-(8)), the number of atoms here actually is the number of beads, we have made related supplementary description in the manuscript.

The second question is meaningful, which is closely related to some subjects. Temperature is a physical quantity that represents the degree of heat and cold of an object. From the perspective of micro/nano scale, it is the intensity of the thermal motion of the molecules and atoms in the object. In fact, this kind of transformation is a statistical question. Thermodynamics deals with the basic laws governing heat whereas *Statistical Mechanics* 10 explains the laws of thermodynamics in terms of kinetics of atoms or molecules in a macroscopic object. Temperature is a macroscopic quantity, while molecular dynamics simulation can only describe microscopic physical quantities, such as velocity or kinetic energy. Since matter is composed of molecules and atoms, and the kinetic energy of each molecule or atom is constantly changing, the average kinetic energy of all molecules or atoms in the system is the focus of attention, becoming the bridge connecting micro-scale kinetic energy to macro-temperature. In addition, the equilibrium distribution of velocities obeys the Maxwell-Boltzmann law 11. So the temperature transferred from relative kinetic energy is calculated in Eq.(8) , which is actually the real physical temperature. The calculation method has widely used in previous studies 12,13 in friction temperature calculation. **Related edits have been added in the manuscript:**

“The total kinetic energy of atoms (beads) in the self-defined group by gathering all the kinetic energy of every atom (bead):” (**Temperature Calculation section, Paragraph 1, line 3**)

1. **Comment:** The sliding speed (50 m/s) is too fast and unrealistic, compared to real experiments. The authors need to discuss the effect of the unrealistic large speed.
2. **Reply:** Thank you for your rigorous consideration, and we totally understand your concern. In fact, this is the usual parameter setting in MD simulations. Due to the huge difference in the scale of time and space and reality in molecular dynamics, the setting of physical parameters (such as speed, pressure) will be much higher, even several magnitude, such as in Dai et al.’s friction simulation 9, in polymer-on-polymer friction simulation conducted by Sivebaek et al. 14, in Mostafavi’s simulation 15. in Chen et al.’s research 12 et al, where *v* represent the sliding velocity. There is no uniform standard for parameter settings in MD simulations, and they are still huge compared to the actual values. But this does not mean that this setting is unreasonable. Even at such a high speed, the temperature curve in our paper during the polymer friction process can firstly rise to a peak temperature and then fall to converge to a stable value, which is consistent with the temperature trend obtained in previous experiment testing 16. As for the cross-scale problem, MD simulation has always been unavoidable. At present, there is no good measure to perfectly compare the time and space scale in macro and micro/nano scale. Meanwhile, this is also the future research direction of coarse-grained molecular dynamics, mesoscopic molecular dynamics, cross-scale molecular dynamics and other methods, but the difficulty still exists and is limited by computer hardware, force field development and algorithm improvement etc.
3. **Comment:** Previous studies (for example, Dai, et al. ACS Appl. Mater Interfaces 8, 8765 (2016)) have shown that chemical bond breaking and remaking can change the friction and wear mechanisms. In the current consideration, bond breaking was not allowed. Therefore, there were no chemical breaking and remaking processes. How does this assumption affect the conclusions of the present work?
4. **Reply:** Thanks for your paper recommendation. Firstly, the introduction of Dai’s paper also pointed out that the atomic structure of the surface plays a decisive role in the friction characteristics of the amorphous film, which is consistent with the description in our paper. In Dai et al.’s research 17, the formation and breaking of chemical bonds across contacting interfaces was found to be closely related to the wear and friction behavior at the nanoscale.

As shown in our paper, we emphasize the importance of physical behavior on the friction interface such as molecular rearrangement and friction model including SSF and RSF on heat generation during sliding process. The united-atom or coarse-grained MD simulation method we used now is more suitable for physical behavior research without chemical structure details, which has been descripted in previous papers 18,19. Regrettably, in our current research, chemical reaction is hard to show, where there were no actual C or H atoms in our models, which are replaced by simple beads. The chemical mechanism of the friction interface is indeed a research direction. We will consider this issue in the follow-up research. For example, the Reaxff force field can better show the chemical reaction of the interface, which has been used to simulate atomically locked interfaces during mechanical friction 20. **Related edits have been added in the manuscript:**

**“**Additionally, the simulation in our research is more biased towards physical behaviors such as molecular rearrangement on the friction surface. Due to the limitations of simulation methods and force fields, the simulation of bond breaking and bond generation cannot be captured. However, the interface chemical changes during the friction process cannot be ignored. For example, high temperatures could occur at the asperity spot-to-spot contacts during friction, directly leading to tribochemical reactions. In Dai et al.’s research, the formation and breaking of chemical bonds across contacting interfaces was found to be closely related to the wear and friction behavior at the nanoscale. Further studies about chemical effect on friction process needs to be further investigated.**”**(**Discussion section, the last Paragraph**)

1. **Comment:** Due to the above issues, there are some uncertainties over the simulation results and conclusions. There is strong need for the authors to provide certain validations (either experimental and theoretical works). However, the authors did not present any comparisons and validation for their results. This is a significant concern.
2. **Reply:** Thank you for your suggestions. In response to your comments above, we have made all the explanations and corresponding amendments, hoping to solve some of your doubts about our research. Based on the concern of lacking corresponding validations, we have added theoretical and experimental verification in the suitable position of the paper. **Related edits have been added in the manuscript:**

“The temperature continues to be close to the quenched layer (), which was considered as a important temperature point that play a significant role in following friction heating process based on our previous studies.” (**Initial Conditioning section, Paragraph 1, Fourth-to-last line**)

“The temperature trend follows Michaeli et al.’s result, who measured the temperature between two polypropylene (PP) granules friction interface by using temperature sensors. Dropped temperature indicates that less and less mechanical energy is dissipated in the latter stage of friction.” (**Heat Generation section, Paragraph 1, line 8**)

**“**Additionally, the simulation in our research is more biased towards physical behaviors such as molecular rearrangement on the friction surface. Due to the limitations of simulation methods and force fields, the simulation of bond breaking and bond generation cannot be captured. However, the interface chemical changes during the friction process cannot be ignored. For example, high temperatures could occur at the asperity spot-to-spot contacts during friction, directly leading to tribochemical reactions. In Dai et al.’s research, the formation and breaking of chemical bonds across contacting interfaces was found to be closely related to the wear and friction behavior at the nanoscale. Further studies about chemical effect on friction process should be mentioned.**”**(**Discussion section, the last Paragraph**)

“In addition, this difference of heat generation in two processes can also be explained from the perspective of bulk temperature and flash temperature. In Ashby et al.’ s research, bulk temperature (mean or bulk surface temperature due to uniform friction surface) and flash temperature (local temperature due the rough surface contact) was analyzed over a wide range of load and sliding velocity. Compared with bulk temperature, flash temperature is more higher, and the level of needed external pressure and velocity is lower. Our simulation result is qualitatively in agreement with their conclusions. In RSF, molecular chains over friction surface are always in a disordered state, making friction interface in a dynamic rough state, where the flash temperature dominates the heat generation process. In contrast, the high degree of orientation caused by SSF leads to a more uniform contact, in which and the body temperature causes heat generation. The different heat generation mechanism also indicates the significance of molecular rearrangement on heat generation.” (**Discussion section, the last paragraph**)

1. **Comment:** A surface temperature of a sliding couple over a wide range of load and sliding velocity was studied before (see Ashby et al. Tribology Tran. 34(4), 577 (1991)). Some discussions and comparisons with this work might be insightful.
2. **Reply:** Thank you for your recommendation. We have read the literature carefully, and made corresponding analysis and comparison in our research paper. **Related edits have been added in the manuscript:**

“In addition, this difference of heat generation in two processes can also be explained from the perspective of bulk temperature and flash temperature. In Ashby et al.’ s research, bulk temperature (mean or bulk surface temperature due to uniform friction surface) and flash temperature (local temperature due the rough surface contact) was analyzed over a wide range of load and sliding velocity. Compared with bulk temperature, flash temperature is more higher, and the level of needed external pressure and velocity is lower. Our simulation result is qualitatively in agreement with their conclusions. In RSF, molecular chains over friction surface are always in a disordered state, making friction interface in a dynamic rough state, where the flash temperature dominates the heat generation process. In contrast, the high degree of orientation caused by SSF leads to a more uniform contact, in which and the body temperature causes heat generation. The different heat generation mechanism also indicates the significance of molecular rearrangement on heat generation.” (**Discussion section, the last paragraph**)

Special thanks to you for your good comments.

**Reviewer #2**

**General Comments:** The manuscript titled “Progressive Molecular Rearrangement and Heat Generation of Amorphous Polyethene Under Sliding Friction: Insight from the United-Atom Molecular Dynamics Simulations” mainly studied the interfacial friction heating mechanisms of PE under the Single Sliding Friction (SSF) and Reciprocating Sliding Friction (RSF) by molecular dynamics (MD) simulations, respectively. The results showed that the RSF process exhibited a better heat generation performance than SSF process, which was tightly related to the molecular motion. Generally, the work is interesting and well written, contains interesting results for the theoretical and experimental science community. However, some issues listed below should be addressed before consideration of publication.

1. **Comment:** There are some grammatical errors in the manuscript. For instance, in Introduction section, Paragraph 2, line 8, “what factor mainly influence” should be rewritten as “what factor mainly influences”?
2. **Reply:** Thank you for your reminder. We have checked out the whole paper carefully, and made due grammatical changes. **Related edits have been added in the manuscript:**

“Molecular dynamics (MD) simulations is considered as a reliable way to study the surface

deformation of the polymer friction under nanoscale.” (**Introduction section, Paragraph 2, line 1**)

“But the question still exist: what factor mainly influences the heat generation of the sliding friction of polymers” (**Introduction section, Paragraph 2, line 8**)

“loading equilibrium stage and friction stage with applied external pressures and velocity” (**Loading and Friction Equilibrium section, Paragraph 1, line 1**)

“and the way that subtracting the influence of external velocity was used when calculating temperature.” (**Temperature Calculation section, Paragraph 1, line 2**)

“Where is the vector between atom (i) and atom (i+10)” (**Chain Orientation and Entanglement Parameter Analysis section, Second-to-last line**)

“At a low pressure,” (**Initial Conditioning section, Paragraph 1, line 13**)

“The rougher surface was obtained in RSF than that in the SSF process” (**Molecular Rearrangement section, Paragraph 3, Third-to-last line**)

**Similar grammatical errors have been modified in the manuscript, and marked in yellow.**

1. **Comment:** Some variables are not in italics, like the number of atoms k and n. “Ebond” in the left side of Eq. 1 should be “Etotal”. The center-of-mass kinetic energy below Eq. 3 should be (Ek)cm, not (Ek)total. The authors should proof-read the manuscript to minimize grammatical errors.
2. **Reply:** Thank you for your suggestion,and we have checked out the whole paper carefully, and made due grammatical changes. **Related edits have been added in the manuscript:**

* **Equation part**

“” (**Interaction Potentials and Modeling System section, Equation 1**)

“” (**Temperature Calculation section, Paragraph 2, line 1**)

* **Italic variable part**

**The following variables in manuscript have been revised to italics, and marked in yellow:**

Glass transition temperature (); Loading pressure (***P***); Velocity parameter (***V***); Amplitude (***A***); Frequency (***f***); Number of atoms such as ***k***, ***n***, and the id of atoms including ***i.***

“ axis”(**Molecular Rearrangement section, Paragraph 3, line 4 and line 6**)

1. **Comment:** The authors give the information of the model and simulation process, however, some details like the equilibrium process and model generation should be provided for others to follow their work. In addition, the author did not give the density of the amorphous PE after equilibrium. Is it comparable with that in experiment?
2. **Reply:** Thank you for your suggestion. In order to make readers better imitate our work, we have made the corresponding supplement. In fact, the simulation result is slightly lower than the experimental value for low density PE (), where was obtained, we deem this acceptable for the united atom method potential employed, which previous works 3,5 have also claimed. **Related edits have been added in the manuscript:**

“A fixed layer was used to avoid atoms exceed the boundary during loading and friction. Quenched layers are maintained at a constant temperature to dissipate the heat generated during sliding. Atoms in the free layer can move freely due to force generated by the potential between atoms. External velocity was applied on the slider, and pressure was applied on the rigid layer meanwhile during friction simulation. To maintain the density of initial layer structure, walls were set on the specific position in the *Z* direction.” (**Interaction Potentials and Modeling Systems section, Paragraph 2, line 5**)

“Canonical ensemble (NVT) was used to regulate the system temperature, relaxation was completed in the isothermal-isobaric (NPT) ensemble. The model was first equilibrated with a temperature of 500 K. NPT ensemble was applied at a temperature of 500 K, and with a external pressure of 1 bar for 300 ps. In the subsequent cooling process, temperature was decreased from 500 K to 100 K over 500 ps (, , and cooling rate of ); the specific volume information of amorphous PE every 100 timesteps were collected during the cooling process.” (**Glass Transition Temperature section, Paragraph 1, line 5**)

“In addition, higher temperature results in a lower density of the PE bulk system in the relaxation process, for example, at 100 K and at 300 K, which is close to the Bao, Hossain et al.’sd result. However, the simulation result slightly lower than the experimental value for low density PE (), which contains a high degree of short and long chain, and on the another hand, we deem this acceptable for the united atom method potential employed.” (**Glass Transition Temperature section, Paragraph 1, line 15**)

1. **Comment:** The authors used “PE block” as “PE bulk” in the manuscript. Maybe the authors should unify them for clarity.
2. **Reply:** Thank you for your reminder,we are sorry for our wrong writing. For better clarity, we have uniformly changed “PE block” to “PE bulk”, which have been marked in yellow in the manuscript.
3. **Comment:** There are many fitting curves in the manuscript. However, the color of the fitting curves should better be same as that of the corresponding simulation data. For example, in Figure 3B, the color of the fitting curve under a pressure of 0.8 MPa is same as that the simulation data under a pressure of 0.2 MPa. This might make the readers confused.
4. **Reply:** Thank you for your suggestion, and for better readability, we have modified the following figures in manuscript.



**Abstract Graph** in Manuscript (**Page 35**)



**Figure 3** in Manuscript (**Page 11**)



**Figure 4** in Manuscript (**Page 12**)



**Figure 6** in Manuscript (**Page 15**)

1. **Comment:** In Heat Generation section, line 16, what’s the meaning of “friction temp” ?
2. **Reply:** Thank you for your suggestion. Friction temp here actually represents the interfacial temperature during friction.To avoid readers’ doubts, we have done some changes in manuscript. **Related edits have been added in the manuscript:**

“Because no obvious temperature gradient in *Z* direction was found in our research due to a thin thickness of polymer. Hence, temperature or frictional temperature calculated here can be approximatively regarded as the interfacial temperature during friction.” (**Heat Generation section, Paragraph 1, line 4**)

“The temperature curve with a lower sliding is rather stable.” (**Heat generation section, Paragraph 1, line 20**)

1. **Comment:** The velocity of the simulations is extremely high. Actually, the loading rate of MD simulation in other researches is also extremely high indeed. I am just curious about it.
2. **Reply:** We totally understand the reviewer’s concern. In fact, this is the usual parameter setting in MD simulations. Due to the huge difference in the scale of time and space and reality in molecular dynamics, the setting of physical parameters (such as speed, pressure) will be much higher, even several magnitude, such as in Dai et al.’s friction simulation 9, in polymer-on-polymer friction simulation conducted by Sivebaek et al. 14, in Mostafavi’s simulation 15. in Chen et al.’s research 12 et al, where *v* represent the sliding velocity. There is no uniform standard for parameter settings in MD simulations, and they are still huge compared to the actual values. But this does not mean that this setting is unreasonable. Even at such a high speed, the temperature curve in our paper during the polymer friction process can firstly rise to a peak temperature and then fall to converge to a stable value, which is consistent with the temperature trend obtained in previous experiment testing 16. Actually, the loading pressure of 0.2, 0.4 and 0.8 MPa is the pressure setting in LAMMPS MD package, representing the added force on every atom of the group, which means every bead in the rigid layer in our paper. If these pressures correspond to the pressure of the entire rigid body layer, it must be multiplied by the number of beads in the rigid body layer. For example, 0.2\*3600=0.72Gpa, 0.4\*3600=1.44Gpa, 0.8\*3600=2.88Gpa. For macro cognition, it is also a huge number, which is also inevitable for MD to study the micro environment. The real area of contact difference between micro/nano scale and macro scale leads to the result that the mean contact pressure in MD can be as large as 3 orders of magnitude larger than the normal applied pressure 1,2. In the past similar friction process under pressure loading using MD method, the pressure value is almost the order of GPa. So in terms of parameter setting, my pressure value is no problem. To facilitate readers’ understanding, **related supplementary explanations have been added in the manuscript:**

“and normal loading pressure *p* (0.2, 0.4 and 0.8 MPa) was applied to every beads in the rigid body, if transferred to the whole rigid layer, it will be the pressure of *n* (number of beads of the group) *p* (, , .). Due to the real area of contact difference between micro/nano scale and macro scale, the mean contact pressure in MD can be as large as 3 orders of magnitude larger than the normal applied pressure.” (**Loading and Friction Equilibrium section, Paragraph 1, line 5**)

1. **Comment:** In Figure 11, the authors analyzed the distribution of bond length in different times. However, the change of the bond length is not obvious in Figure 11A.
2. **Reply:** Thank you for your suggestion. It is really true as you pointed that the quantitative change is not obvious, but the bond length trend as described in the manuscript can greatly follow both the energy and temperature trend curve. And the reason why this change is not obvious can be owning to the interatomic potential we use in our research, which do not allow dynamic bond formation or scission. **Related edits have been added in the manuscript:**

**“**Additionally, the simulation in our research is more biased towards physical behaviors such as molecular rearrangement on the friction surface. Due to the limitations of simulation methods and force fields, the simulation of bond breaking and bond generation cannot be captured. However, the interface chemical changes during the friction process cannot be ignored. For example, high temperatures could occur at the asperity spot-to-spot contacts during friction, directly leading to tribochemical reactions. In Dai et al.’s research, the formation and breaking of chemical bonds across contacting interfaces was found to be closely related to the wear and friction behavior at the nanoscale. Further studies about chemical effect on friction process should be mentioned.**”**(**Discussion section, the last Paragraph**)

Special thanks to you for your good comments.

At last, we gratefully thank the editor and all reviewer for their time spending making their constructive remarks and useful suggestions, which has significantly raised the quality of the manuscript. We tried our best to improve the manuscript and made some changes in the manuscript.These changes will not influence the content and framework of the paper. And here we did not list the changes but marked in yellow in revised paper. We appreciate for Editors/Reviewers’ warm work earnestly, and hope that the correction will meet with approval.

Once again, thank you very much for your comments and suggestions.

Thank you and best regards.

Yours sincerely,

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