Molecular Dynamics Simulations of Contact Electrification in Natural Rubber Polymer for Triboelectric Nanogenerators

การจำลองพลวัตเชิงโมเลกุลของการเกิดประจุไฟฟ้าที่ผิวสัมผัสในพอลิเมอร์ยางธรรมชาติสำหรับการประยุกต์ใช้งานเป็นแหล่งกำเนิดไฟฟ้านาโนแบบไทรโบอิเล็กทริก

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ABSTRACT

We use reactive molecular dynamics (MD) simulations in conjunction with charge equilibration method to investigate the atomic-scale mechanism of contact electrification between polyisoprene and copper compared to that between polyisoprene and polytetrafluoroethylene (PTFE) for the application of triboelectric nanogenerators (TENGs). Our atomistic simulations show that the natural rubber polymer can either gain or lose electrons when it is in physical contact with copper or PTFE. In addition, the charge transfer at the polyisoprene-PTFE interface is greater than that of the polyisoprene-copper system govern by the energetics of a partial charge transfer reaction, which could provide an understanding of the underlying mechanism responsible for enhancement of the TENG performance.

บทคัดย่อ

งานวิจัยนี้ใช้การจำลองพลวัตเชิงโมเลกุลร่วมกับวิธีสมดุลประจุเพื่อศึกษากลไกระดับอะตอมของการเกิดประจุไฟฟ้าจากการสัมผัสระหว่างคู่วัสดุพอลิไอโซพรีนและทองแดง เปรียบเทียบกับการสัมผัสระหว่างคู่วัสดุพอลิไอโซพรีนและพอลิเตตระฟลูออโรเอทิลีน เพื่อการประยุกต์ใช้เป็นเครื่องกำเนิดไฟฟ้านาโนแบบไทรโบอิเล็กทริก ผลการจำลองระดับอะตอมแสดงให้เห็นว่าพอลิเมอร์ยางธรรมชาตินี้ สามารถเป็นได้ทั้งตัวรับหรือสูญเสียอิเล็กตรอนเมื่อเกิดการสัมผัสกับทองแดงหรือพอลิเตตระฟลูออโรเอทิลีนตามลำดับ นอกจากนี้เราพบว่า การถ่ายโอนประจุที่ผิวสัมผัสระหว่างพอลิไอโซพรีนกับพอลิเตตระฟลูออโรเอทิลีนนั้น มีค่ามากกว่าการถ่ายโอนประจุระหว่างพอลิไอโซพรีนกับทองแดง ทั้งนี้ถูกควบคุมโดยแรงขับเคลื่อนทางพลังงานไฟฟ้าจากปฏิกิริยาการถ่ายโอนประจุ โดยการศึกษานี้ สามารถช่วยเพิ่มความเข้าใจเกี่ยวกับกลไกพื้นฐานในการเพิ่มประสิทธิภาพของเครื่องกำเนิดไฟฟ้านาโนแบบไทรโบอิเล็กทริกได้

Keywords: Molecular dynamics (MD) simulation, Triboelectric nanogenerator, Contact electrification
คำสำคัญ: การจำลองพลวัตเชิงโมเลกุล เครื่องกำเนิดไฟฟ้านาโนแบบไทรโบอิเล็กทริก การเกิดกระจุไฟฟ้าด้วยการสัมผัส

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Introduction

Renewable energy, which is inexhaustibly produced from natural sources such as solar energy, water energy, and wind energy, has become increasingly popular in recent years as it is expected to replace petroleum, facing the crisis due to large global demands. Normally, electrical energy is consumed and converted to mechanical energy such as the movement of the machinery driven by electricity. According to the principle of energy conservation, mechanical energy can also be converted back to electrical energy, and there is a kind of renewable energy that utilizes the physical movements occurring in our everyday life. Despites its merits, this energy source had been neglected so far. Until a recent decade, the devices converting mechanical energy into electrical energy, the so-called triboelectric nanogenerators (TENGs), have attracted much attention as promising alternative energy sources.

TENGs was discovered by Zhong et al. (2012) at Georgia Institute of Technology. It is the technology that has been developed to produce electricity from mechanical energy derived from human or environmental movements, such as the movement of vehicles, the flow of water and wind. A contact of two materials exhibiting different ability to gain or lose electrons results in charge transfer at the interface based on the principle of contact electrification (Zheng et al., 2017). For example, rubbing an amber stick against a wool and placing it near a wood chip causes the two materials to be attracted to each other due to an electrostatic force originating from the charge transfer between materials. Each material has different tendency to gain or lose electron when it is in physical contact with the others as mentioned, and this behavior is characterized according to the triboelectric series (Molnar et al., 2018; Lee et al., 2018). Specifically, the magnitude of charge exchange between two materials is determined by their relative position in the triboelectric series. Based on this principle, Zhong Lin Wang et al. have created 4 different types of TENGs, including 1) vertical contact-separation mode, 2) contact-sliding mode, 3) single-electrode mode, and 4) freestanding triboelectric-layer mode. From this development, TENGs have been expected to be applied for a wide variety of devices such as the electricity generated by knee or elbow bending (Yi et al., 2015), by touching between foot and ground, or by touching between hand and doorknob (Wang et al., 2017).

Enhancement of the TENGs performance can be achieved by appropriately selecting a pair of materials according to the triboelectric series. Researchers have also explored new materials to increase the efficiency of TENGs, including oil-resistant buna-N rubber and food-grade oil-resistant buna-N/vinyl rubber, which are positively charged when they are in contact with liquid mercury (Zou et al., 2019). In addition to finding the new materials, material mixtures and composites have also been examined, for example, silver particles were added to natural rubber to improve the triboelectric properties (Appamato et al., 2021).

Despites significant progress in TENGs development, the atomic-scale mechanisms governing contact electrification phenomena are difficult to be examined using solely experimental techniques, and computer simulation methods have been adopted as a tool for investigating the atomistic behaviors of materials. Yoshida et al. used density functional theory (DFT) to study the mechanisms of charge transfer between aluminum and polytetrafluoroethylene (PTFE) (Yoshida et al., 2006). They found that the amount of charge transfer is negligible when the orientation of PTFE molecules is parallel to an aluminum slab. For the perpendicular case in which a dangling bond of the polymer chains is adjacent to the metal, significant amount of charge transfer was observed. Similar results have been reported by Wu et al. who studied the contact electrification phenomena in a PTFE-Al couple using a DFT method (Wu et al., 2018). Charge transfer between aluminum and PTFE with a dangling bond at the end of the molecular chain was compared to that between the metal and the polymer that does not have the defect. It was found that the defect can promote the amount of charge transfer, governed by a lower interface energy barrier. Moreover, they also found that the amount of charge transfer increases with the decrease of the interface distance. Although this high-level first principles calculation results in high accuracy, the simulation size and time scale are limited. Using classical molecular dynamics (MD) simulation method can capture a longer time scale and larger simulation size, which could provide additional insights into the unanswered questions regarding the charge transfer mechanisms.

In this research, we aim at answering what governs the direction and amount of charge transfer between materials and how molecular structure of materials affects their electrification property. We use reactive MD simulations, which is capable of capturing bond breaking and formation, to study the electrification process in polyisoprene when it is in physical contact with copper and PTFE. These two contact materials were chosen since they are located above and below polyisoprene in the triboelectric series, respectively. Polyisoprene was selected as our prototypical case because it is a main component of natural rubber, which is one of the main economic products of Thailand. Accordingly, the material with a good quality is readily available and cheap. Natural rubber can easily be formed into various shapes as it is very flexible. Moreover, it is lightweight, chemical resistant, and can be used in the temperature range of -55 to 70°C. This material is consequently suitable for utilizing in TENG applications (Appamato et al., 2021).

Objectives of the study

1. To study the atomic-scale mechanism of charge transfer in polyisoprene using reactive molecular dynamics simulation in conjunction with charge equilibration method.
2. To compare the amount of charge transfer between polyisoprene-copper and polyisoprene-PTFE (Polytetrafluoroethylene) systems.
3. To understand the driving force and energetics of contact electrification that govern the charge transfer phenomena.

Methodology

Molecular dynamics (MD) simulation and charge equilibration (QEq) method

To describe the interactions between atoms in this work, we used molecular dynamics (MD) simulations with reactive force field (ReaxFF), which has an ability to capture bond breaking and bond formation using the relationship among bond order, bond distance, and bond energy. The total interaction energy in ReaxFF consists of bond, valence-angle, torsion-angle, lone-pair or coordinates, under coordinate, van der Waals, and Columbic energy terms given by the following equation:

$$E_{system} = E_{bond} + E_{val} + E_{tor} + E_{over} + E_{under} + E_{lp} + E_{vdw} + E_{coulombic} \tag{1}$$

The ReaxFF force field parameter adopted in this study follows the work of Islam (Islam et al., 2017). We also compared the reliability of the ReaxFF force field with non-reactive force field. All molecular dynamics simulations were performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code (Plimpton, 1995).

To study the charge transfer phenomena at a junction between materials, we used charge equilibrium method following the work of Rappe et al. (Rappe et al., 1991). Considering a system with N atoms possessing an individual charge of Q_i, a total electrostatic energy of the system can be written as:

$$E_Q(Q_1 \dots Q_N) = \sum_A (E_{A0} + \chi_A^0 Q_A) + \frac{1}{2}\sum_{A,B} Q_A Q_B J_{AB} \tag{2}$$

where E_{A0} is the energy of atom A at a neutral state, χ_A^0 is the electronegativity of atom A given by

$$\chi_A^0 = \left(\frac{\partial E_A}{\partial Q_A}\right)_{A0} \tag{3}$$

and J_{AB} is the Coulombic interaction between the charged atom A and B calculated from the second derivative of energy with respect to charge as follows:

$$J_{AB} = \frac{\partial^2 E_A}{\partial Q_B^2} \tag{4}$$

From this total electrostatic energy function, charge distribution optimization can be calculated using the conditions that all atomic chemical potentials are equal:

$$\chi_1 = \chi_2 = \cdots = \chi_N \tag{5}$$

and a total charge is equal to the sum of all partial charges:

$$Q_{tot} = \sum_{i=1}^{N} Q_i \tag{6}$$

Systems of interest and simulation of contact electrification

We used the Polymer Modeler tool on nanoHUB (Haley et al., 2020) to construct the molecular structure of polyisoprene. Generally, polymers (in a solid phase) have a molecular weight as high as ~10^4-10^6 g/mol; however, simulation of the extremely large molecule is computationally expensive. Thus, we first find the optimal molecular size to reduce a computational cost while natural properties of the material are still maintained. To do this, 6 polyisoprene samples with various numbers of molecular chains and different numbers of repeat units per chain were created; all have the same total number of repeat units of ~400. These include the systems containing 1) 40 molecular chains each comprising 10 repeat units, 2) 20 chains each consisted of 20 units, 3) 13 chains each having 30 units, 4) 10 chains with 40 units per chain, 5) 8 chains each consisted of 50 units, and 6) 4 molecular chains with 100 repeat units per chain. Each system was first subjected to an equilibration simulation using the DREIDING force field with fixed atomic charges generated from the nanoHUB tool. To this end, the isobaric-isothermal (NPT) simulations at an atmospheric pressure and a room temperature have been performed for 1 ns using a timestep of 1 fs. Then, the ReaxFF simulations in conjunction with charge equilibration method were performed for the obtained structures. The simulations have been carried out for 100 ps under the NPT ensemble using a timestep of 0.1 fs. Temperature and pressure were controlled using the Nosé-Hoover thermostat and barostat with coupling constants of 100 and 2,500 fs, respectively.

The molecular structure of polyisoprene obtained from the simulation is shown in Figure 1a and 1b, where white, gray and brown colors denote H, single-bond C and double-bond C atoms, respectively. Figure 1c compares density as a function of molecular weight of polyisoprene obtained from RexaFF (purple symbol) and non-reactive (green symbol) MD simulations. We observe for the low regime (below ~2,750 g/mol) that the density increases with increasing molecular weight. The density of polyisoprene remains almost unchanged for the molecular weight ≥ 2,750 g/mol corresponding to a number of repeat units of 40. Therefore, the system containing 10 molecular chains with 40 repeat units per chain was chosen for our production simulations. When comparing the simulated density of polyisoprene of this molecular size, ReaxFF simulation gives rise to a value of 1.01 g/cm^3, significantly higher than the non-reactive simulation result with a value of 0.88 g/cm^3. However, the density of polyisoprene derived from ReaxFF force field is comparable to the experimental observation (0.92 g/cm^3), indicating that the implemented method is suitable for the study.

After a force field validation step and finding an optimal molecular size, we performed the simulations to investigate charge transfer behavior at the polyisoprene-copper and polyisoprene-PTFE interfaces. To do this, a polyisoprene slab with free surfaces along the z-direction, containing 10 chains each comprising 40 repeat units was constructed. The slab was subjected to 100 ps-equilibration simulation under the canonical (NVT) ensemble with a temperature of 300 K to relax the free surfaces. Similarly, copper and PTFE slabs were prepared; the PTFE system is consisted of 3 molecular chains each having 10 repeat units. The well-equilibrated copper and PTFE slabs were attached to the polyisoprene, accompanied by additional NVT simulations for 100 ps to equilibrate the systems.

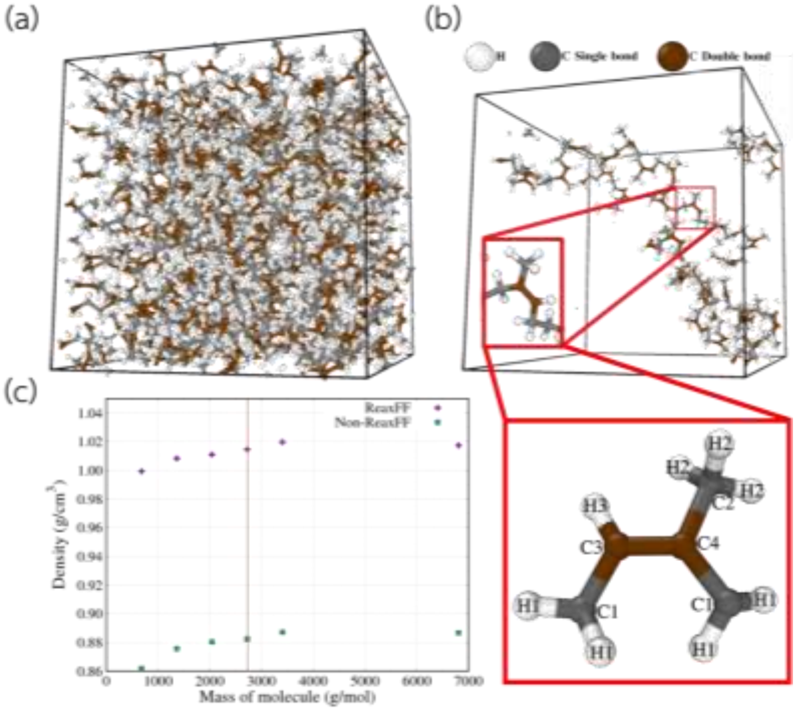

Figure 1 (a) Atomistic snapshot of a simulation cell containing 10 polyisoprene molecular chains each comprising 40 repeat units. (b) A single molecular chain and a single repeat unit of polyisoprene. White, gray and brown spheres represent H, single-bond C and double-bond C atoms, respectively. (c) Density as a function of molecular weight.

Results and Discussion

Figure 2 displays atomistic snapshots from ReaxFF MD simulations of (a) polyisoprene-copper and (b) polyisoprene-PTFE contact interfaces. Color code of the snapshots on the left panel is represented based on atom type, while that on the right denotes atomic charge, i.e., red, green, and blue represent positively charged, neutral, and negatively charged atoms, respectively. Evidently, the atoms located in vicinity of the interfaces have distinguishable charge color compared to the other regions, indicating that charge transfer event primarily occurs at the contact interface. The right panel of Figure 2a illustrates that the interface copper atoms become positively charged as they appear in red color, while the atoms of polyisoprene in this region (in light-blue color) are negatively charged. On the other hand, the polyisoprene has an opposite charge when the material is attached to PTFE as demonstrated on the right panel of Figure 2b. The interface PTFE atoms are clearly visible as dark blue color, suggesting that they gain electrons from the polyisoprene. Our simulation results qualitatively show the direction of charge transfer between the materials in good agreement with the triboelectric series from experimental observation (Molnar et al., 2018).

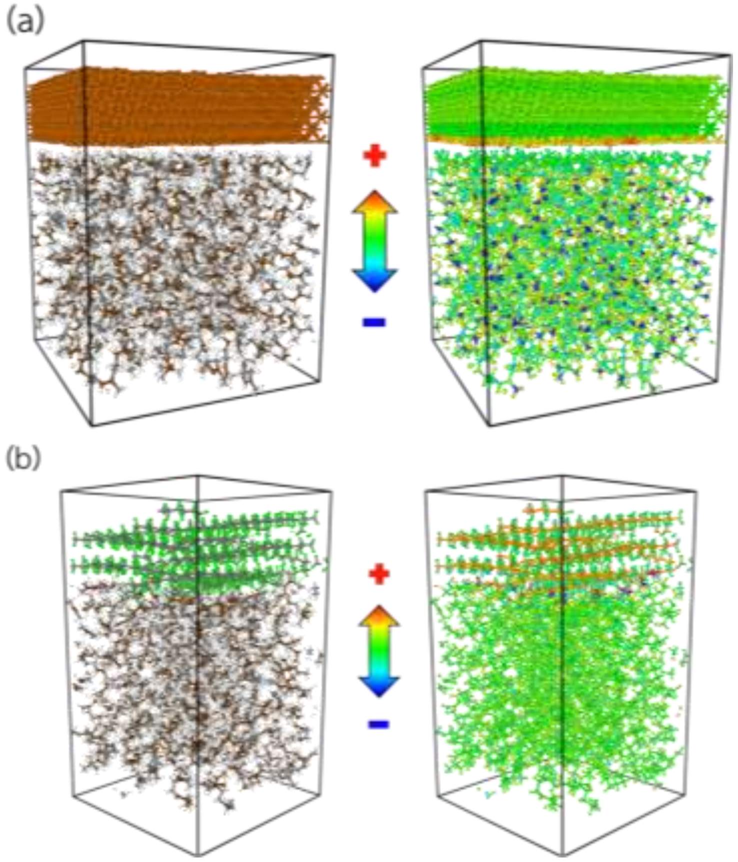

Figure 2 Atomistic snapshots from reactive MD simulations of (a) polyisoprene-copper and (b) polyisoprene-PTFE interfaces. The left panel displays atomic color based on atom type, where white, gray, brown, and green represent H, single-bond C, double-bond C, and F atoms, respectively, while the atoms displayed on the right panel are colored based on atomic charge: red–positively charged, green–neutral, and blue–negatively charged.

To quantitively analyze the charge transfer at the interface between materials, we calculate charge density profiles of the systems. To do this, the simulation cells were divided into small bins along the z-direction, and charge density of each bin was computed by summing all atomic charges in a particular bin divided by a cross-sectional area of the simulation box. Figure 3a displays charge density as a function of the position along the z-direction of polyisoprene-copper (red) and polyisoprene-PTFE (blue) junctions. In this figure, the interface position was set at zero, and the negative and positive distance from the interface represent the regions of copper (or PTFE) and polyisoprene, respectively. It is clearly seen that charge transfer event mainly occurs at the interfaces as we observe the peaks at a position of below ~4 Å, while the rest part of polyisoprene located at a farther distance remains almost neutral. Our quantitative analysis also reveals that polyisoprene can either gain or lose electrons depending on the material it is attached to. The material become negatively and positively charged when it is in contact with copper and polyisoprene, as observed in Figure 3a. This is in accordance with the results from atomistic snapshots shown in Figure 2. In addition to the direction of charge transfer, we also compared a magnitude of charge accumulated at the interface of polyisoprene. For this analysis, we integrated charge density profiles (Figure 3a) from a distance of zero (the interface position) to ~4 Å (the end of charge density peak), and the results are presented in Figure 3b. By comparing the magnitude of charge, we found that the induced charge density at the polyisoprene-PTFE interface of 170 mC/m^2 is greater than that at the polyisoprene-copper junction, $|-125|$ mC/m^2. In other words, the amount of charge per area that polyisoprene loses to PTFE is higher than that it gains from the copper. This is corresponding to the difference in a relative position between materials in the triboelectric series, which governs charge transfer effectiveness.

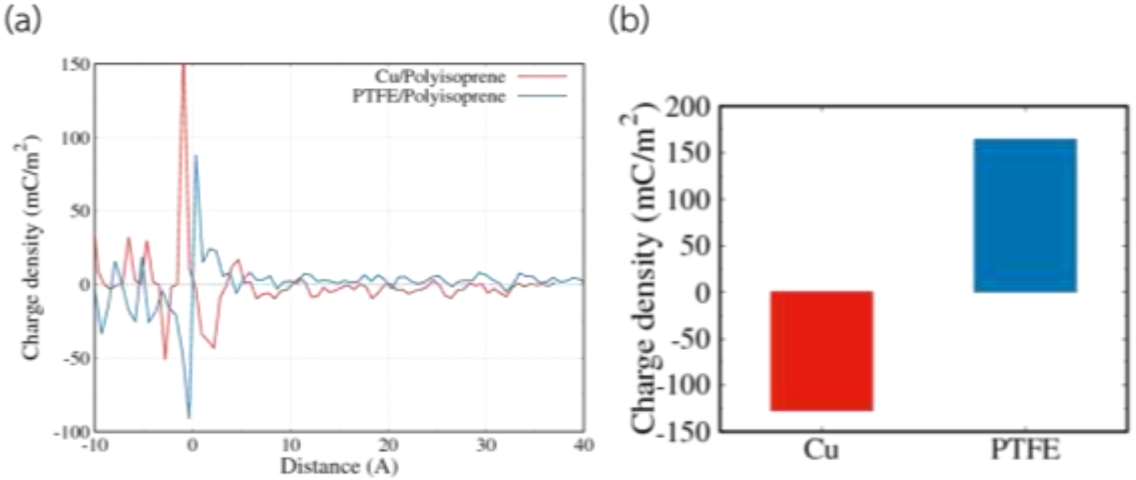

Figure 3 (a) Charge density profiles of polyisoprene-copper (red line) and polyisoprene-PTFE (blue line) systems. (b) Charge density accumulation at interfaces of polyisoprene attached to copper (red bar) and PTFE (blue bar).

As discussed above, our simulations yield the results regarding the direction and magnitude of charge transfer at the interfaces of materials in good agreement with experiments. While the previous analysis provides a macroscopic behavior of charge transfer between materials comparable to the trends predicted using the triboelectric series, a microscopic view, in particular the atomic-scale charge evolution, has not been discussed yet. Next, we are interested in examining the atomic charge variation in the molecular structure of polyisoprene. Hereof, seven types of atoms in the repeat unit of polyisoprene, including C1, C2, C3, C4, H1, H2, and H3, were categorized as shown in Figure 1. Figure 4 shows average partial charges of each atom type in polyisoprene before (red bars) and after (blue bars) attaching to copper (4a) and PTFE (4b). Purple bars in Figure 4 represent the difference between these quantities, which can be used to indicate the local sites mainly involved in charge transfer event. It is shown that the partial charges of C2 and H2 atoms are dominantly altered, while that of C1 and C3 remain almost unchanged, which could be attributed to a strong covalent interaction at this local sites. For the polyisoprene-copper junction shown in Figure 4a, although the C2 atom becomes less negatively charged after a slab attachment, the reduced positive charges of hydrogen atoms overall results in electron gain in polyisoprene. The other way around has been observed for the case of polyisoprene-PTFE junction as displayed in Figure 4b. Taking all into account, our simulations suggest that the dangling $-CH_3$ unit in polyisoprene structure (see Figure 1) plays a key role on the contact electrification behavior of the material. This could provide an insight into molecular design for enhancing the charge transfer between materials in TENGs.

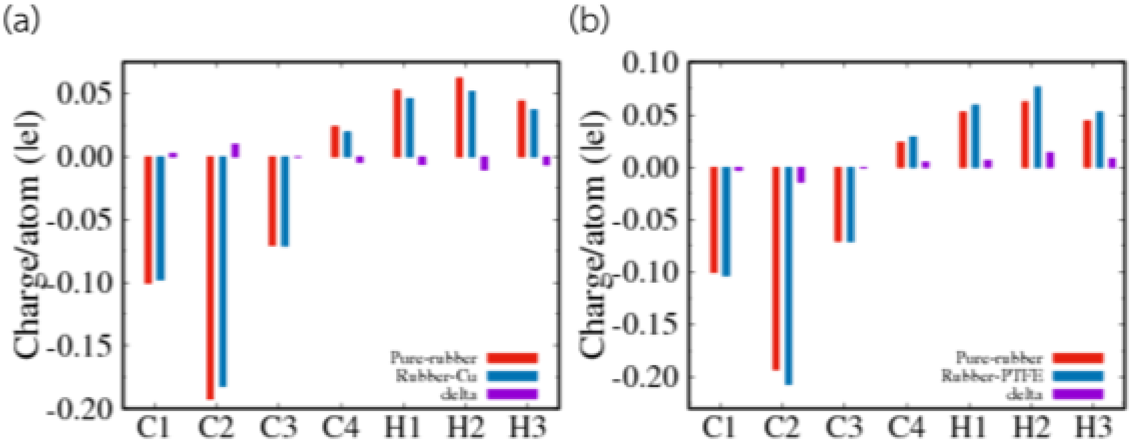

Figure 4 Average partial charges of various atom types in polyisoprene structure for (a) polyisoprene-copper and (b) polyisoprene-PTFE systems. Red, blue, and purple bars represent the quantity before contact, after contact, and their difference, respectively.

Finally, we are interested in investigating the energetics of a contact electrification reaction to understand what the driving force governing charge transfer between materials is. We calculated the total electrostatic energy given by Equation 2 for various systems displayed in Figure 5a, including copper (E_{copper}), polyisoprene ($E_{polyisoprene}$), and polyisoprene-copper ($E_{polyisoprene\text{-}copper}$). Then, formation energy (E_f) was computed using Equation 7. The same applies for the polyisoprene-PTFE system.

$$E_f = E_{polyisoprene-copper} - (E_{polyisoprene} + E_{copper}) \quad (7)$$

The calculated formation energy is negative for both systems as demonstrated in Figure 5b, indicating the energy released from the formations of these interfaces. Comparing polyisoprene-PTFE (blue bar) with polyisoprene-copper (red bar) system, we found that the polyisoprene-PTFE interface formation is thermodynamically more stable as it exhibits more negative formation energy. In addition, the charges averaged throughout polyisoprene system induced by the two materials it is in contact with were calculated as illustrated in Figure 5c. Similar to charge density analysis in Figure 3b, the magnitude of the transferred charge per atom in polyisoprene-PTFE (blue bar in Figure 5c) is greater than that in the polyisoprene-copper (red bar in Figure 5c). Following the concept of the electronegativity given by Equation 3, which is the rate of electrostatic energy change with respect to charge, we calculated the formation energy per unit charge transferred as shown in Figure 5d. This quantity indicates the amount of energy released per a unit of charge gain or loss, which can imply the thermodynamics of the reaction. Figure 5d illustrates that the magnitude of the quantity: $\frac{dE}{dQ}$ of polyisoprene-PTFE (blue bar) is higher than that of the polyisoprene-copper system (red bar). This implies that the reaction involving a unit charge transferred from polyisoprene to PTFE is energetically more favorable than the material gaining a unit charge from copper. Overall, our simulations reveal atomic-scale mechanisms of charge transfer between materials in which the amount of charge transfer depends on the driving force or the energetics of contact electrification.

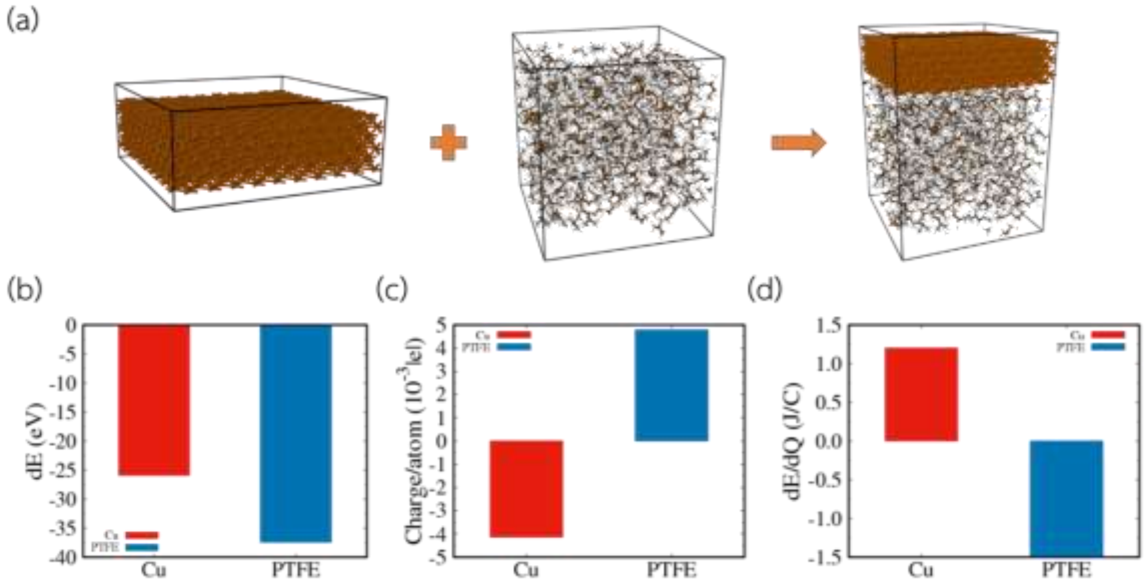

Figure 5 (a) Atomistic snapshots of copper slab, polyisoprene slab, and polyisoprene-copper junction system for electrostatic energy calculations. (b) Formation energy, (c) charge per atom, and (d) formation energy per a single unit of charge transfer in polyisoprene. Red and blue bars represent the quantities for polyisoprene-copper and polyisoprene-PTFE system, respectively.

Conclusions

In conclusion, we used reactive molecular dynamics simulation with charge equilibration method to study charge electrification phenomena in polyisoprene induced by copper compared to polytetrafluoroethylene (PTFE). We found that polyisoprene can either gain or lose electrons depending on the material it is attached to. The material becomes negatively and positively charged when it is in contact with copper and PTFE, respectively. Moreover, the amount of charge transfer between polyisoprene-PTFE is greater than that between polyisoprene-copper. The obtained results regarding the direction and magnitude of charge transfer are in good agreement with the macroscopic behaviors whose trends can be predicted using the triboelectric series. The analysis of charge gain or loss in a particular atom type constituting a molecular structure of polyisoprene suggests that the dangling –CH_3 unit plays a key role on charge exchange. Moreover, the energetics of charge transfer reaction has also been examined, and we found that the energy released per a single unit of charge gain or loss is the driving force governing the amount of charge transfer between materials. This study can provide mechanistic insights into contact electrification phenomena, beneficial for enhancing the performance of triboelectric nanogenerators.

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