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Article

The Adsorption and Conductivity of Silicon in Elastic Polymers

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Abstract: Silicon (Si) is the major material of semiconductor element. For semiconductor element producing, Si need to refined into high-purity silicon crystals, which has the approximate characteristics of intrinsic semiconductor, then doping 13th group or 15th group trace element. Producing the high-purity silicon crystals with well-lattice structure need a lot of energy and highly cost. Besides, the high-purity silicon is very hard and fragile which limits the scope of application. For this reason, this study added low-grade non-complete lattice silicon in the elastic polymer, and then doped with high concentrations of fine carbon powder. This material can be conductive by physisorption; therefore, it can be used to making semiconductors. Using this material to produce electronic components can significantly reduce the cost of purification and crystallization, saving energy at the same time. Moreover, because this elastic polymer materials containing silicon, which has soft, warm, and durable physical properties. With those properties, the design of the device could be more flex, full scalability and close to the skin.

Keywords: elastic polymer; adsorption; conductivity

1. Introduction

Conducting polymers have improved the sensitivity of a wide variety of sensors due to their excellent electrical conductivity or the charge transfer properties [1]. Conducting polymers hold the advantages of promise as flexible, inexpensive materials for using in electronic applications including solar cells, light-emitting diodes, and chemiresistor-type sensors [2–4]. Therefore, a simple, scalable, cost-effective deposition technique for conducting polymers that produces a uniform thin film morphology reproducibly is needed [5].

In this study, the electrical characteristics of elastic silicon polymers were investigated, like resistance temperature, conductivity etc. The results showed that elastic silicon polymers can broadly applied to producing heating device, temperature control systems, sensors, variable resistors and other related fields.

1.1. Absorption and Conductivity

Polymers can be deposited in a desired location on any surface, such as film, glass, and metal [6]. After heating the elastic polymer material which doped both carbon and silicon, the low melting point polymer will completely dissociated and evaporated, leaving high melting point carbon and silicon particles. The quality and size of those particles are the same as before. Therefore, there is no chemical reaction between the carbon, silicon and elastic polymers, but only physisorption. Physisorption is causing by the pore size of carbon and the intermolecular attraction can occur in any kinds of material. However, the adsorptions of each material are very different. For that reason, when carbon adsorb with different chemical composition in elastic polymers will have different resistivity.

Adsorption by porous adsorbents is a possible means of removing some of the volatile toxicants from smoke [7]. Despite the litany of reported methods including electrostatic adsorption [8], their limitations range from reproducibility issues and cost effectiveness to lack of scalability. Therefore, a universal solution is needed that reliably deposits any conducting polymer film on any substrate [5]. The experimental results showed that when the elastic polymer containing silicon will significantly reduce the resistance and thus increase the conductivity of elastic polymer.

1.2. The Characteristics of Silicon in Polymer

Organic silicon polymer is polymer containing silicon atoms in the molecular structure, usually refers to silicones. Silicones are a polymer with table structure which alternating composed by silicon atoms and oxygen atoms, the basic unit is —Si—O—. The silicon atoms also connect with variety of organic functional groups, such as methyl, ethyl and phenyl etc. Those compounds are semi-organic and semi-inorganic polymers. The oxygen plasma can promote the $-O_nSi\ (OH)_{4-n}$ group on the silicon surface, which is highly dependent on the intensity of oxygen plasma and the treatment time [9].

The chemical structure of the silsesquioxane family is defined as $R_nSi_nO_{1.5n}$, which makes a structure consisting of an inner inorganic framework of silicon and oxygen atoms, externally covered by organic groups (R) [10]. R may either be hydrogen or an alkyl, alkene, aryl, and arylene group. Based on their molecular architecture, silsesquioxanes can be classified into two main categories of noncaged and caged nanostructure. The noncaged type silsesquioxane molecules can be further classified into random, ladder, and partial-cage nanostructure [11]. The ladder like polysilsesquioxanes which include poly (phenyl silsesquioxane), poly (methyl silsesquioxane), and poly (hydrido)silsesquioxane [12–14]. The enormous importance of organosilicon compounds in both polymer and synthetic organic chemistry provided satisfactory methods for the installation of the dimethylsilanol unit through: (1) addition of organometallic compounds to silicon electrophiles followed by simple or oxidative hydrolysis, (2) transition metal catalyzed hydrosilylation, and (3) silyl insertion [15–19]. Silanols are not well known as reagents in organic synthesis but they are air and water stable reagents that can be chromatographed on silica gel and distilled [20].

2. Materials and Methods

2.1. Elastic Silicon Polymers

Because organic silicon polymers are semi-organic and semi-inorganic polymer, they have a lot of excellent performance, like high temperature tolerance, unaging, insulation, hydrophobia, fireproofing, chemically inert, some silicon polymers can even resist corrosion, and electromagnetic wave. The performance of silicon polymers is much better than others, therefore silicon polymers can widely use in electronics, chemicals, machinery, construction, textile, transport, medicine and other fields [21,22]. According to the above, this study produced semiconductors by doping high concentrations of graphite into silicone polymers, hope it can widely use electronic in technology. Here we develop this phenomenon into a solution-based method to grow films of nanostructured conducting polymers that could facilitate their use in applications ranging from actuators to sensors [23,24].

2.2. The Structure of Elastic Silicon Polymers

The structure of elastic silicon polymers related to the chemical bond, molecular structure and polymer structure etc. For facilitate follow-up study, this research use scanning electron microscope (SEM) to analyze the polymers structural and bonding structure.

2.3. SEM Analysis

The molecular packing for polymer has major impact on its physical property. The carbon physisorption reduces the polymer intermolecular distance and increases largely the intermolecular attraction. Adsorptions for different doping density of polymeric rubberlike material are observed by SEM electron, and images are shown on the Figures 1–4. Figure 1 is SEM image of silicon polymers doping with 0M graphite. Figure 2 is SEM image of silicon polymers doping with 4M graphite. Figure 3 is SEM image of silicon polymers doping with 50M graphite. Figure 4 is SEM image of polyurethane rubber doping with 75M graphite. SEM analysis showed that the graphite particles are independent particles, the average particle size of graphite particles is 5μm, no chemical reaction between any particles.

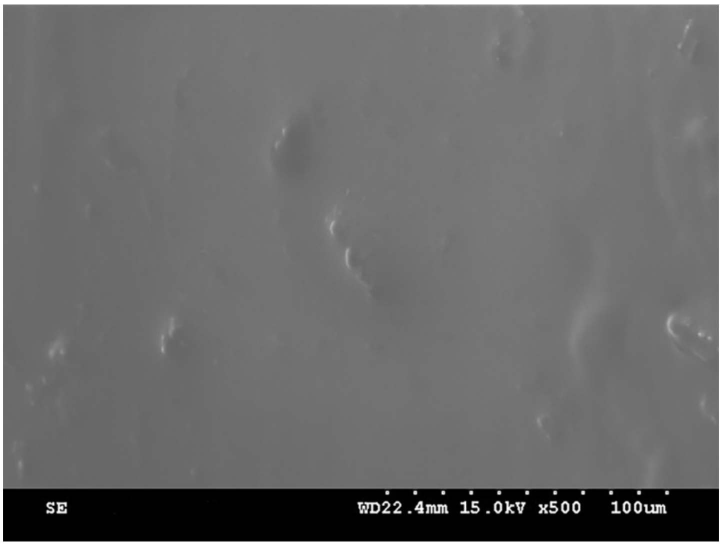

Figure 1. SEM image of silicon polymers doping with 0M graphite.

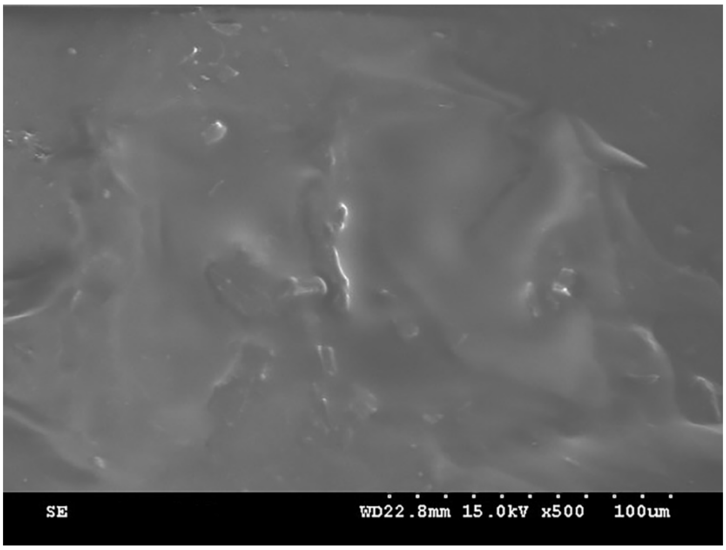

Figure 2. SEM image of silicon polymers doping with 4M graphite.

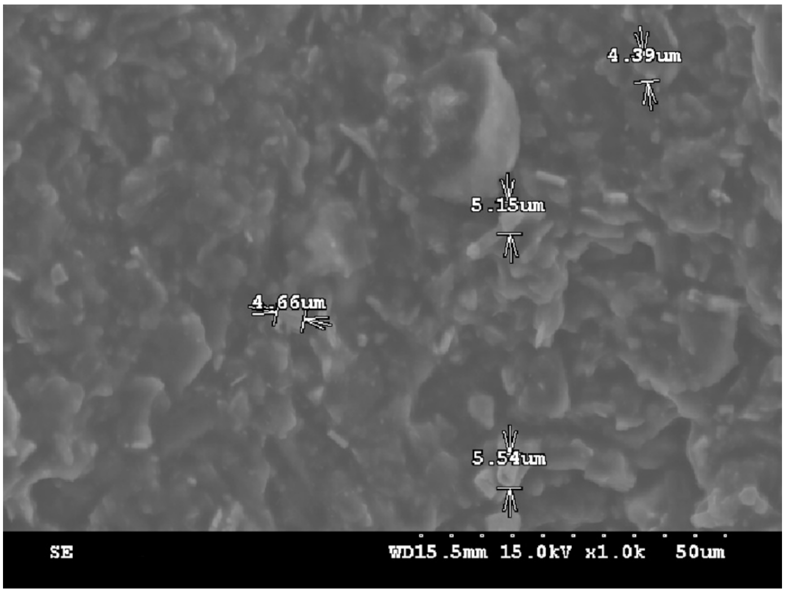

Figure 3. SEM image of silicon polymers doping with 50M graphite.

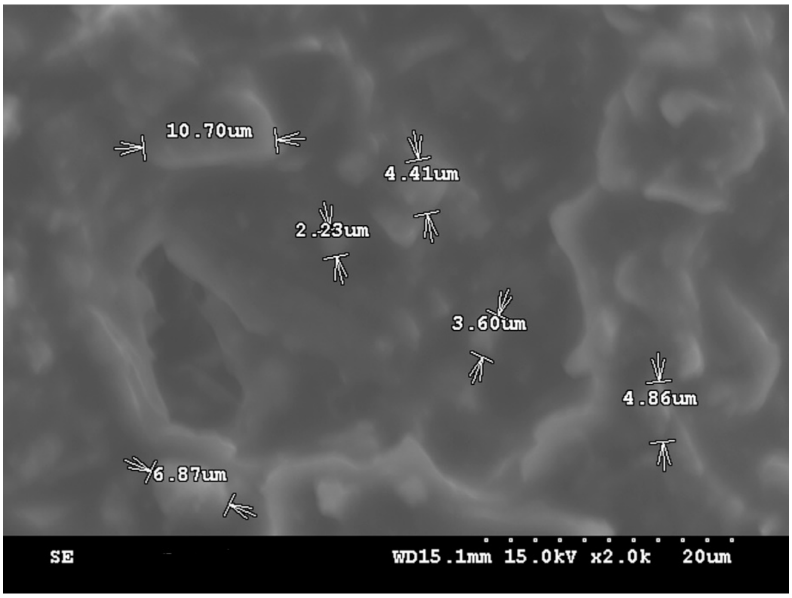

Figure 4. SEM image of silicon polymers doping with 75M graphite.

The polymer second level of key knot strength and macromolecular weight will cause the intramolecular high-residual force. High carbon doping density has enormous attractions to make molecular pile up closely, and presents high regularity structure. Since the polymer has huge molecular weight and structure flaw, it cannot be arranged in parallel.

2.4. EDX Analysis

This study using energy dispersive X-ray (EDX) to investigate the composition and content of polymers specimen. Figures 5, 6 and Table 1 are the EDX analysis results. Those results showed that the silicon polymers specimen contain 13% silicon after 50M graphite doped, suggest that silicon atomic structure highly affects the adsorption and conductivity of polymers.

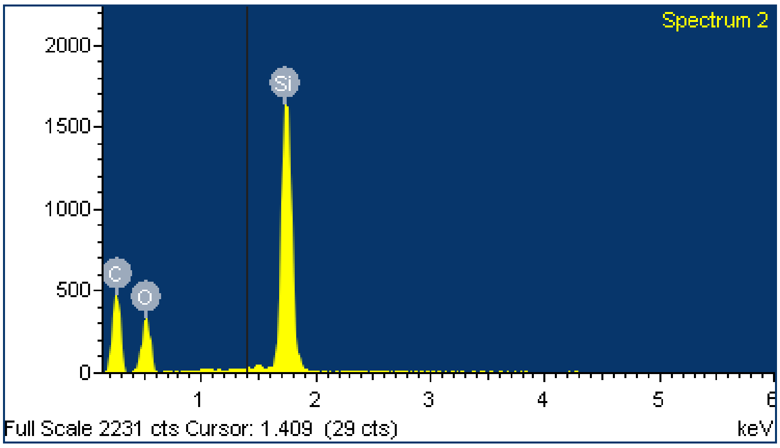

Figure 5. EDX image of silicon polymers doping with 50M graphite.

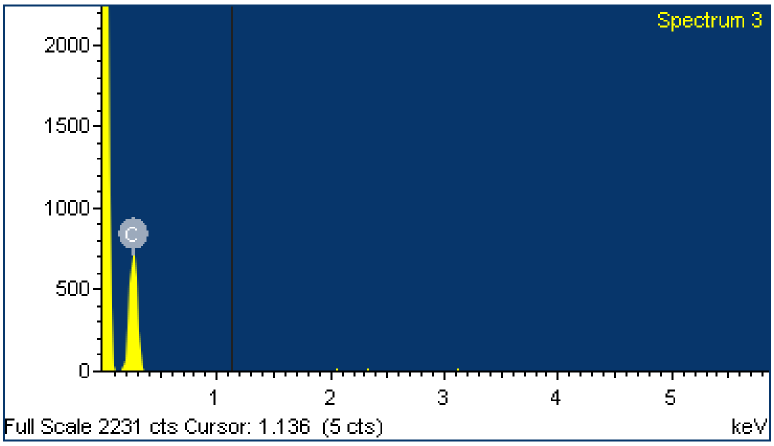

Figure 6. EDX image of polyurethane rubber doping with 75M graphite.

Table 1. The EDX analysis result. (A) Silicon polymers doping with 50M graphite specimen (B) Polyurethane rubber doping with 75M graphite specimen.

Sample	**A**		**B**	
Element	Weight%	Atomic%	Weight%	Atomic%
C	49.64	62.58	95.29	96.42
O	25.21	23.86	4.71	3.58
Si	25.15	13.56		
S				
Totals	100		100	

2.5. The Infrared Absorption Spectrum Analysis

The infrared absorption of organic silicon compounds is 5 times stronger than the hydrocarbons. The infrared absorption spectrum of organic silicon compounds is varied at range 1290 cm^{-1} to 800cm^{-1}. Table 2 shows the characteristic absorption peaks of major functional groups. Figure 7 is the infrared absorption spectrum image of silicon polymers doping with 50M graphite. Figure 8 is the infrared absorption spectrum image of Polydimethylsiloxane.

Table 2. The characteristic absorption peak of organic silicon compounds.

Chemical groups	Peak / cm^{-1}	The characteristics of peak
Si—H	2250~2100 950~800	Si—H Extension vibrating absorption, strong Si—H Bending vibrating absorption, strong
Si—C	890~690	Si—C Extension vibrating absorption, strong

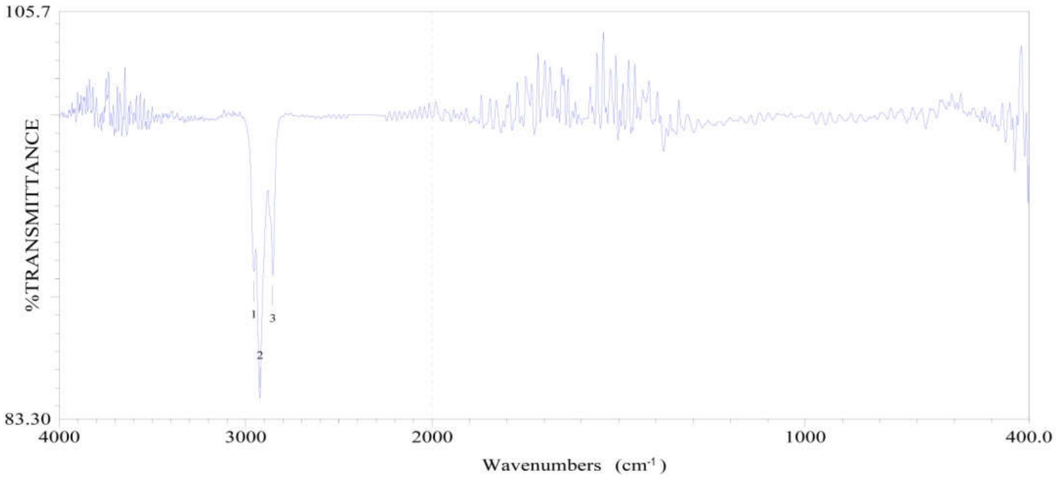

Figure 7. The infrared absorption spectrum image of silicon polymers when it doping with 50 M graphite.

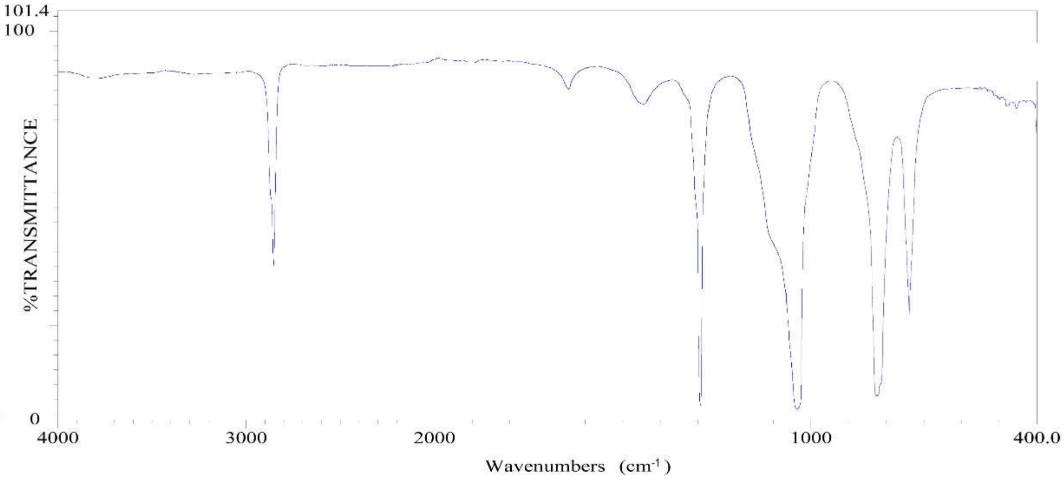

Figure 8. The infrared absorption spectrum image of polydimethylsiloxane [$Me_3Si (OSiMe_2)_n OSiMe_3$], degree of polymerization n=0.

2.5.1. The Infrared Spectra of Silicon-Oxygen Bond (Si-O)

According to Figure 6, Si-O bond have a strong peak in zone 1100~1000cm^{-1}, this peak coursing by the anti-symmetric extending vibration of the Si-O-Si bond. To linear polysiloxane, with the growth of chain the peak divided into two similar intensity peaks, which are located at 1085cm^{-1} and about l020cm^{-1}. Cyclic polysiloxane is also absorbed in the same area. The Si-O band of trimeric ring siloxane $(R_2Si)_3$ shows an extension vibrating absorption peak at 1020cm^{-1}. The absorption peak of tetramer ring or the bigger ring moved to 1090cm^{-1}. The substituents binding with silicon can also affect the position of Si-O-Si bond absorption peak. For example, Cl_3Si-O-$SiCl_3$ and Si-O-Si absorption located at 1124cm^{-1}.

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2.5.2. The Infrared Spectra of Silica

Comparing Figures 5 and 6, the infrared spectra analysis of silica, showed that although the silicon polymers doping with graphite, there have no Si-O bond and Si-C bond. Suggest that there has no chemical reaction between silicon and graphite particles, they are completely independent.

3. Results

3.1. Conductive

For investigating the electrical characteristic of different concentration carbon was doped in silicon. Elastic polymers were produced by silica, which has high absorbability to carbon, with different concentration of graphite doped. To further improve detection, other functionalities were employed to increase analyte binding [25].

Figure 9 shows the resistivity (ϱ) of specimen with different concentration (C_D) carbon doped. When the elastic polymers contained 40M graphite, the conductive will increase rapidly. When the elastic polymers contained 50M graphite, the conductive get into steady state. When the elastic polymers contained 60M graphite, graphite adsorption reached to saturation. The flexibility of the polymer began to deteriorate, so the test materials have tiny cracks in the edge.

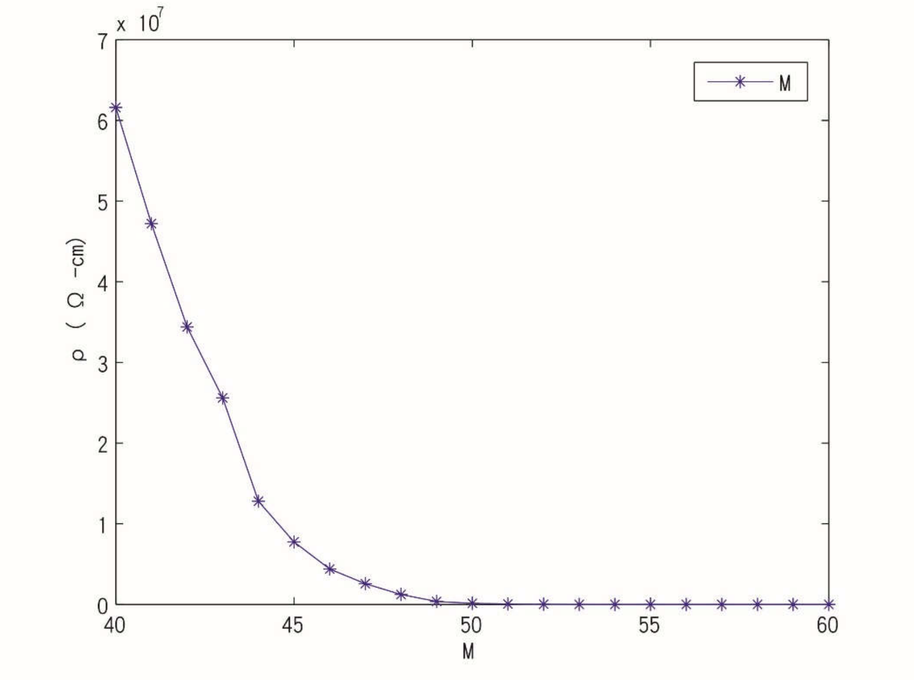

Figure 9. The resistivity (ϱ) of specimen with different concentration (C_D) carbon doped.

For investigating the adsorption and conductive of silicon, elastic polymers were produced by polyurethanes rubber, which has high absorbability to carbon and no silicon contained, with different concentration of graphite doped as control. Figure 10 shows the resistivity (ϱ) of polyurethanes rubber with different concentration (C_D) carbon doped. When the elastic polymers contained 75M graphite, the conductive of elastic polymers increased rapidly. When the graphite increases to 79M, the conductive of elastic polymers get into steady state. When the elastic polymers contained 90M graphite, graphite adsorption reached to saturation. The flexibility of the polymer began to deteriorate, so the test materials have tiny cracks in the edge.

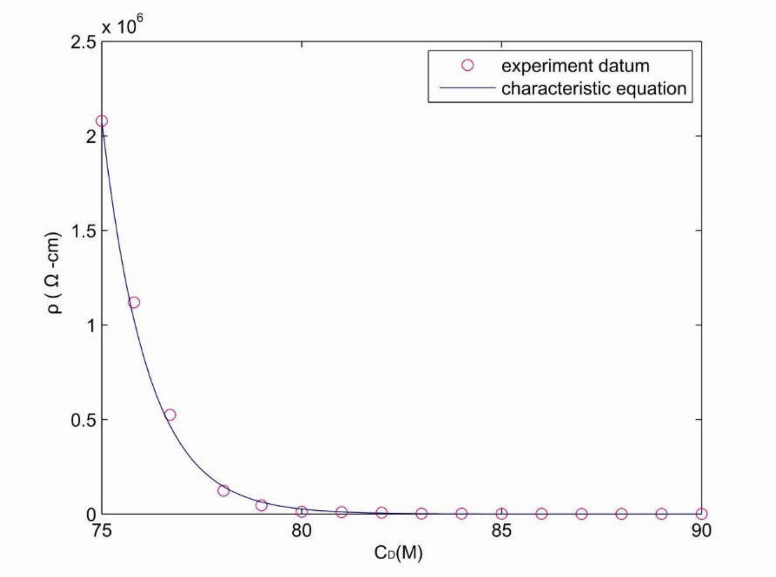

Figure 10. The resistivity (ϱ) of polyurethanes rubber with different concentration (CD) carbon doped.

3.2. Resistance of Temperature Coefficient

Theoretical studies have determined that the energy required for removing adsorbed particles from any interface is much greater than the energy required for spreading [26]. Physisorption is not a chemical reaction, needed no activation energy, therefore it can react easily and quickly respond at low temperature. Molecular interactions between the free surface energy of interfacial adsorbed nanofibers and the substrate can dictate film morphology [27]. Adsorption is exothermic, so the resistance, which caused by the physisorption of polymer, proportional to temperature. The weak Van der Waals force is the main force of physisorption. The physisorption easily affected by temperature, so the resistance of temperature coefficient varies greatly. Because the conductive of elastic polymers contain silicon have great variances with the elastic polymers only, this study analyzed the resistance of temperature coefficient under similar resistivity, but different concentration graphite doped. The result as follow: When the polyurethanes rubber doping whit 46.3M graphite, the characteristics of resistance temperature as Figure 11. When the silica doping whit 60M graphite, the characteristics of resistance temperature as Figure 12.

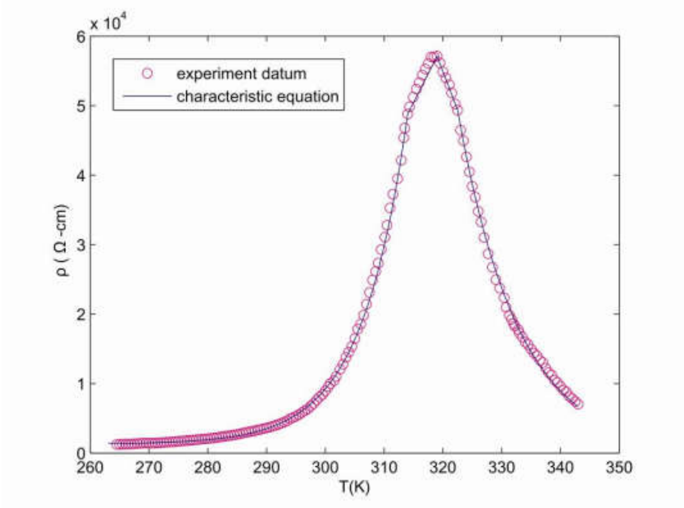

Figure 11. The characteristics of resistance temperature, when polyurethanes rubber doping whit 46.3M graphite.

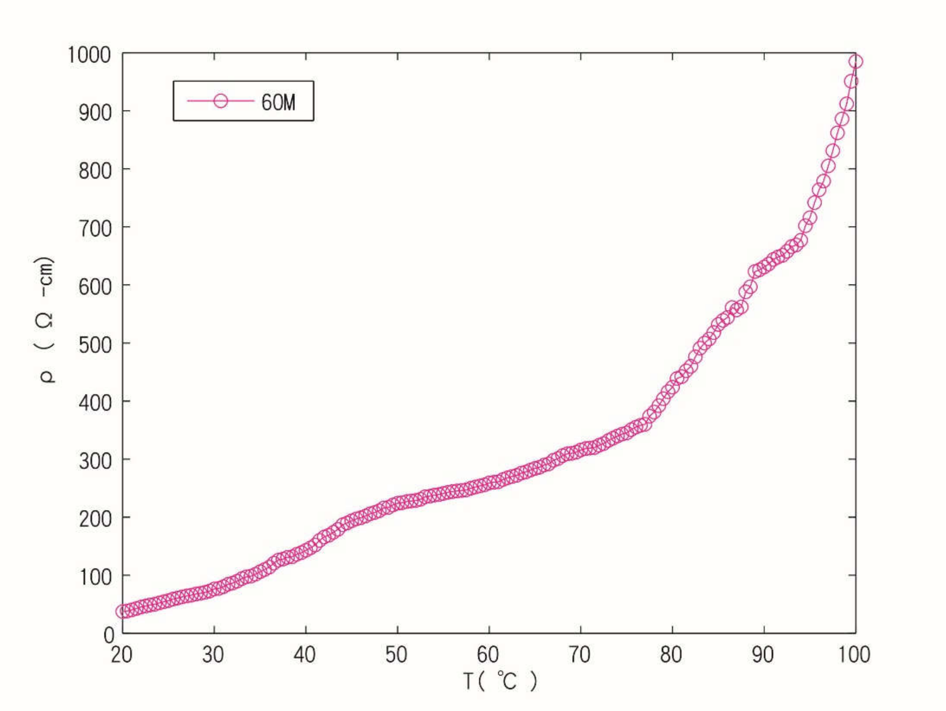

Figure 12. The characteristics of resistance temperature, when silica doping whit 60M graphite.

This experiment uses a nonelectrolyte solution that is not easily volatile. Only the solvent is volatile, and there is no interaction between the solute and the solvent, which is close to an ideal solution and applicable to Raoult's law. According to Raoul's law, the total vapor pressure of a solution is equal to the sum of the vapor pressures of the solute and solvent. If two ideal solutions are mixed together with a constant volume, they will not absorb or release thermal energy. Mixing two solutions and increasing their volume is an endothermic reaction. On the contrary, mixing two solutions reduces the volume, which is an exothermic reaction. Therefore, this experiment is an exothermic reaction.

4. Discussion

4.1. Conductive

When doped 50M graphite into elastic polymers which contain 13.56% silicon, the physisorption resulting the elastic silicon polymers become conductive. The resistivity of the elastic silicon polymers is 140kΩ-cm and its conductivity is 7.1428μS-cm. Add the same concentration of graphite into the elastic polymers contain no silicon, like polyurethane rubber. The resistivity of the polyurethane rubber is 60kΩ-cm and its conductivity is 0.01666μS-cm. Dependent on the infrared absorption spectrum analysis, these two polymers were mainly composed by C-H bond. The variance of conductive between two polymers are 430 times, only because the chemical reaction of silicon particles.

4.2. Temperature Coefficient

Adsorption of the chemisorptions is stronger than physisorption, not easily affected by temperature. The elastic silicon polymers are working by physisorption, easily affected by temperature.

4.3. The Characteristic Equation of Carbon Concentration and Resistance

Through several experiments, derived the characteristic equation of graphite concentration and resistance as follows:

$$\varrho = \varrho_s + \varrho_0 \cdot e^{-Q_m \frac{C_D - C_0}{C_c - C_0}} \tag{1}$$

In Formula (1), C_0 is the lowest doping concentration for using polymers as practicality resistance material; ϱ_0 is the resistivity of the lowest doping concentration; C_C is the doping critical concentration of the steadying conducting state; C_S is the doping saturation concentration of saturated adsorption; ϱ_S is the resistivity of saturation concentration in saturated adsorption; Q_m is the quality factor of material, which is dimensionless. The Q_m of graphite is 4. When temperature raise to 300k, the C_0 of polyurethanes rubber which doping with graphite, is 75M, the C_0 of silica is 45M, the C_C of polyurethanes rubber is 80M, the C_C of silica is 50M, the C_S of polyurethanes rubber is 90M, the C_S of silica is 60M, the ϱ_0 of polyurethanes rubber is $2.08\times10^6\Omega$-cm, the ϱ_0 of silica is $6.16\times10^7\Omega$-cm, the ϱ_s of polyurethanes rubber is $3.04\times10^2\Omega$-cm, the ϱ_s of silica is $5.60\times10^2\Omega$-cm. When C_D is smaller than C_C, Formula (1) can simplify as follows:

$$\varrho = \varrho_0 \cdot e^{-Q \frac{C_D - C_0}{C_c - C_0}} \tag{2}$$

4.4. The Characteristic Equation of Resistance and Temperature Coefficient

Physisorption is not a chemical reaction, needed no activation energy, therefore it can react easily and quickly respond at low temperature. Adsorption is exothermic, so the resistance, which caused by the physisorption of polymer, proportional to temperature. The weak Van der Waals force is the main force of physisorption. Conservation of energy dictates that electron transfer between an electrode and a redox system in solution or adsorbed on the electrode surface is fastest when the energy of the electron is equal in the metal and in the thermally activated redox system [28,29]. The physisorption easily affected by temperature, so the resistance of temperature coefficient varies greatly. When the polyurethanes rubber doping whit 80M graphite, the characteristics of resistance temperature as Figure 9. The characteristic equation of resistivity (ϱ) and temperature as follows:

$$\rho = \begin{cases} \rho 0 \text{, if Te} < \text{T0} \\ \rho 0 + (\rho p1 - \rho 0) \cdot e^{Q_{Td} . D_m \frac{(T_e - T_0)}{T_{p1} - T_0}} \text{, if Te} < \text{Tp1} \\ (\rho c - \rho pl) \dfrac{T_c - T_e}{T_c - T_{p1}} \text{, if Tpl} < \text{Te} < \text{Tc} \\ (\rho nl - \rho c) \dfrac{T_{nl} - T_e}{T_{nl} - T_c} \text{, if Tc} < \text{Te} < \text{Tnl} \\ \rho c \cdot e^{-Q_{Td} \frac{T_e - T_c}{T_m - T_c}} \text{, if Te} > \text{Tc} \end{cases} \tag{3}$$

In Formula (3), T_0 is the temperature of starting desorption; ϱ_0 is the resistivity at T_0, T_{pl} is the starting temperature of positive temperature coefficient linear region, ϱ_{pl} is the resistivity at T_{pl}, Tc is the critical temperature of positive and negative temperature coefficient of rubber polymers, ϱ_C is the resistivity at T_C, T_{nl} is the termination temperature of negative temperature coefficient linear region, ϱ_{nl} is the resistivity at T_{nl}, Tm is the melt temperature of rubber polymers, ϱ_m is the resistivity at T_m, Te is the environment temperature. Q_{Td} is the characteristic of material thermodynamics is dimensionless. The Q_{Td} of graphite is 2. D_m is the characteristic of material desorption, is dimensionless. The D_m of graphite is 2. When polyurethanes rubber doping with 80M graphite, the T_0 =267K, T_{pl} =312K, T_C = 319K, T_{nl} = 322.5K, T_m = 343K, ϱ_0 = $1.296\times10^3\Omega$-cm, ϱ_{pl} = $4.88\times10^4\Omega$-cm, ϱ_C = $5.712\times10^4\Omega$-cm, ϱ_{nl} = 4.936×10^4 Ω-cm , ϱ_m = $7.04\times10^3\Omega$-cm.

When the silica doping whit 60M graphite, the characteristics of resistance temperature as Figure 10. By linear segments can explain the relationship between resistivity (ϱ) and temperature, as piecewise-linear equivalent. Because the linear are close to the actual curve, the characteristic equation as follows:

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$$\rho = \begin{cases} \rho 0 \ \text{, if Te} < \text{T0} \\ \rho 0 + (\rho c1 - \rho 0)\dfrac{T_e - T_0}{T_{c1} - T_0} \ \text{, if T0} < \text{Te} < \text{Tc1} \\ \rho 0 + (\rho c2 - \rho cl)\dfrac{T_e - T_{c1}}{T_{c2} - T_{c1}} \ \text{, if Tcl} < \text{Te} < \text{Tc2} \\ \rho 0 + (\rho m - \rho c2)\dfrac{T_e - T_{c2}}{T_m - T_{c2}} \ \text{, if Tc2} < \text{Te} < \text{Tm} \end{cases} \tag{4}$$

In Formula (4), T_0 is the temperature of starting desorption; ϱ_0 is the resistivity at T_0, T_{cl} is the first section critical temperature of positive temperature coefficient linear region, ϱ_{cl} is the resistivity at T_{cl}, T_{c2} is the second section critical temperature of positive temperature coefficient linear region of rubber polymers, ϱ_{c2} is the resistivity at T_{c2}, T_m is the melt temperature of rubber polymers, ϱ_m is the resistivity at T_m, T_e is the environment temperature. Q_{Td} is the characteristic of material thermodynamics is dimensionless. The Q_{Td} of graphite is 2. D_m is the characteristic of material desorption, is dimensionless. The D_m of graphite is 2. When polyurethanes rubber doping with 60M graphite, the $T_0 = 283K$, $T_{cl} = 350.5K$, $T_{c2} = 368K$, $T_m = 373K$, $\varrho_0 = 1.83\times10^1$ Ω-cm, $\varrho_{cl} = 3.74\times10^2$Ω-cm, $\varrho_{c2} = 7.16\times10^2$Ω-cm, $\varrho_m = 9.85\times 10^2$ Ω-cm.

5. Conclusions

5.1. Silicon Atomic Structure Effect the Adsorption Conductive of Elastic Polymers

Silicon and carbon are located in the same family of the periodic table, their atomic structure and b properties are similar. However, silicon in the third period in 14th families of periodic table, and carbon in the second period, they also have many differences. The atomic radius of silicon is 0.11nm; the volume of silicon is 1.5 times bigger than carbon, so the spacing between silicon and substituent are longer than carbon. For example, the distance between each chlorine atoms within $SiCl_4$ are 0.329nm, and the distance between each chlorine atoms within CCl_4 is 0.298nm. Thus, silicon atoms have poor shielding effect. Therefore, the silicon-containing polymers are much easier hydrolysis than the polymers carbon only.

Adsorption is facilitated by hydrophobic π–π interactions [30]. The silicon atoms are part of a silacyclopentadiene unit, which is responsible for the unique σ^*–π/π^* conjugation that defines their reduced band gap energies [31,32]. Silicon and carbon are reduced π electron shell with the same physisorption, and make the elastic polymers become conductive. Therefore, when adding 50M graphite, the conductive can vary 430-fold between the elastic polymer which contained 13.56% silicon and non-silicon contained. This huge variance of conductive is because of that the electronic structure of silicon has $3s^2 3p^2$ and 3d empty orbital. The 3d electron shell structure can have an empty orbital, can produce dπ-pπ bond, which with the characteristics of quasi-metallic. Carbon atom has no d electrons and no coordination, so when put both carbon and silicon into the elastic polymers; they will adsorb and conducted together which create a better conductive.

5.2. Applicability of The Characteristic Equation

According to experiments and the derivation of characteristic equation, different elastic polymer and containing silicon or not, will not affect the characteristic equation of graphite concentration and resistance, but only change the value of equivalent. Therefore, need more diversity experiments to test that the characteristic equation applies to most of the polymers or not.

5.3. Physisorption Effect the Elastic Polymers

Physisorption can form a single or multiple molecules adsorbed layer, multiple molecules adsorbed layer at the most. Macromolecule and carbon are multiple molecules adsorbed layer, their adsorption rate and rate are very quickly. Therefore, it can reduce the process and production costs.

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