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Relationship between Real Contact Area and Adhesion Force of Plasma-Treated Rubber Sheets Against Stainless-Steel Ball

Jong-Hyoung Kim[1)*], Isami Nitta[2)], Noritsugu Umehara[1)], Hiroyuki Kousaka[1)], Mamoru Shimada[3)] and Mitsuru Hasegawa[3)]

[1)]Department of Mechanical Science and Engineering, Nagoya University
1 Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8603, Japan
[2)]Graduate School of Science and Technology, Niigata University
Ikarashi 2-nocho 8050, Niigata, Niigata 950-2181, Japan
[3)]Nipro Corporation, Research and Development Laboratory
3023 Nojicho, Kusatsu, Shiga 525-0055, Japan
*Corresponding author: hotemail@hanmail.net

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The adhesion force between a chloride-isobutene-isoprene rubber (CIIR) and stainless steel ball was studied in this paper. In order to decrease the adhesion force, the CIIR rubber was treated with high-density microwave plasma employing oxygen and argon gases. The experimental results showed that the adhesion force decreases with increasing treatment time and microwave power following both oxygen and argon plasma treatments. In addition, optical measurements revealed that the real contact area between a CIIR rubber and quartz plate decreases with decreasing adhesion force. The decreasing trend of the contact area was almost similar to that of the adhesion forces. The field emission scanning electron microscope image showed that the surface roughness generally increases with treatment time and microwave power, implying that roughening of rubber surface by plasma treatment is one of the reasons for the decrease in the adhesion force.

Keywords: adhesion force, chloride-isobutene-isoprene rubber, real contact area, plasma treatment, microwave, temperature effect

1. Introduction

The increasing application of polymeric materials has also attracted more attention to the problems associated with their interfacial properties. Their surface properties sometimes do not meet the demands regarding wettability, biocompatibility, gas transmission, adhesion and friction. Especially, the peeling and adhering creates large friction coefficient on the rubber, which leads to wear and subsequent device failure. In order for these rubbers to become more effective in dynamic applications, the surface of the rubber must be modified to reduce adhesion and friction.

A conventional solution is to apply some type of lubrication, like silicon oil, to the surface of the rubber. Depending on the type of lubrication method used, the following problems may arise: (1) the oil may not be permanently bonded to the surface of the rubber and can be wiped away easily (2) the oil can be a source of contamination in medical applications and (3) the rubber seals cannot be re-lubricated in permanently encapsulated devices. Plasma treatment is one of the alternative methods used to overcome the aforementioned problems associated with oil lubricants. It offers an additional advantage that the surface modification does not affect the desirable bulk properties of the rubber. In previous research we discussed about the mechanical properties and interfacial chemistry by the oxygen plasma treatment[1)].

Nano-indentation technique was used to study the mechanical properties in the bulk and subsurface of a CIIR, which is oxygen plasma treated. In particular, the effect of oxygen plasma treatment on the decrease of the adhesion force between CIIR rubber and stainless-steel was discussed. We recognized that oxygen plasma process changed the wettability and surface free energy of CIIR rubber. However, the oxygen plasma treatment of CIIR rubber led to a decrease in contact angle (or an increase in surface free energy) due to an increase in C-O peak. Therefore, surface energy experiment and FTIR analysis results had opposite trend. On the other hand, the Young's modulus of CIIR rubber increased by 38.8% according to the various oxygen plasma treatment conditions. The maximum indentation depth decreased from 400 nm to 360 nm by surface treatment. The mechanical properties have great effect to decrease the adhesive force between CIIR rubber and stainless-steel. Several analytical approaches have been developed to extract mechanical properties by analyzing the nano-indentation load–displacement curve. On the basis of the results of the change of mechanical properties and surface energy, we believe that real contact area could be decreased by the plasma treatment to the rubber. The objective of this research is to find out the relationship between the real contact area and adhesion force of CIIR rubber with plasma treatment. It is also attempted to figure out the effect of temperature on the adhesion force, during oxygen and argon plasma treatments at different time and microwave power.

2. Material and methods

2.1. Material & plasma treatment

The CIIR used contains 98 mass% of isobutene and 1.3 mass% of isoprene. The dimension of a CIIR specimen cut from a CIIR sheet was 3.5×15×30 mm.

Plasma treatment apparatus consisting of a 2.45 GHz microwave generator that can supply power up to 2k W was used in this study. This apparatus is a type of symmetric surface wave-excited plasma source, where high-density columnar plasma along the outer surface of the quartz tube is generated. The generated plasma diffuses to the surface of a specimen 50 mm distant from the quartz surface. Before and after each treatment, the chamber was evacuated to a base pressure below 10^{-3} Pa. The samples were placed in the middle of the chamber. After treatment, the chamber was evacuated to the base pressure again, and then air was introduced into the chamber until atmospheric pressure is reached. Following plasma treatment, all CIIR specimens were stored in closed dishes at room temperature. Four important factors govern the effect of plasma treatment: treatment time, type of gas, gas flow rate, and power input. In this study, we focused on the effect of (1) treatment time with two different gases: oxygen and argon, and (2) power input, on the adhesion forces between a stainless-steel ball and a CIIR specimen treated under various conditions.

2.2. Contact area & adhesion force measurement

After the plasma treatment of a CIIR specimen, pull-off test was conducted to measure the adhesion force between stainless-steel ball and the treated CIIR rubber[2]. For the pull-off test, a SUS 440C (19.05 mm in diameter) was thrust to the CIIR specimen, where a thrust maximum load of 1 N was applied for 90 seconds. All the adhesion tests were experimented in air-conditioned room (22-24 degree, 45-50% RH).

Measurement of real contact area between a quartz plate and a treated specimen was employed with contact microscope[3]. The layout of the contact microscope is shown in Fig. 1. The normal load was applied by

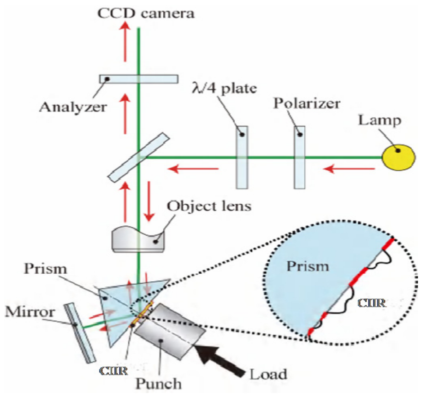

Fig. 1 Schematic diagram of a contact microscope with an elliptically polarized light[3]

turning a blot that pressed the CIIR against the prism. The applied load was measured by a load cell placed between the bolts and the metal base.

After plasma treatment of a CIIR specimen, a column 5mm in diameter and 3.5 mm in height was cut from the treated specimen. The detail of this method is described in ref. 4. Note that all specimens were measured 1-2 days after plasma treatment at room temperature.

2.3. Temperature during plasma treatment

The temperatures of CIIR rubber specimens were measured by a thermocouple which was stuck to the specimen during the plasma treatment. To check the heating effect on the adhesion force of plasma-treated specimen, heat treatment of another fresh specimen was also conducted with an electric furnace and ceramic heating treatment. The heating temperature was set similar to that measured during plasma treatment. Both heating treatments were conducted in atmospheric pressure. In the ceramic heating treatment state, the CIIR rubbers were heated on the heater die. While in the electric furnace condition, we inserted the CIIR rubber into the chamber after the temperatures are reached. After heating treatment, the adhesion forces were measured within 5 minutes.

2.4. Morphology analysis

A Hitach S-570 field emission scanning electron microscope, operated at an acceleration voltage of 15.0 kV, was used to take photomicrographs of the untreated and plasma treated CIIR rubber samples. Before testing, these samples were sputter coated with platinum for 1minute.

3. Results and discussion

Schematic of the real contact areas between a prism and CIIR rubber is shown in Fig. 2. Note that the surface roughness condition would be different from prism and stainless steel ball which is used to measured the adhesion force. However, this study focused on the surface modification such as roughness and real contact

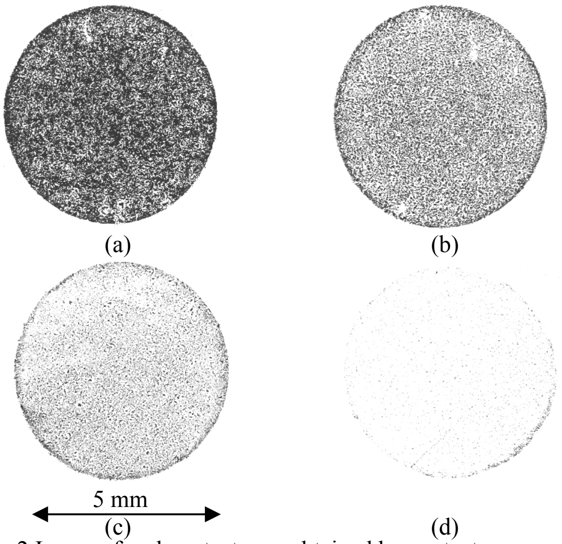

Fig. 2 Image of real contact area obtained by contact microscope for CIIR specimens treated with O_2 plasma at 200 W for
(a) 0 (Untreated) (b) 5 (c) 10 and, (d) 15 min

area change during the plasma treated to CIIR rubber. The applied was 5 N in order to control similar to applied load in the adhesion test by calculating based on the Hertz theory. The appearance real contact area was slightly reduced according to the oxygen and argon plasma treatment time. The longer the time of the oxygen and argon plasma treatment resulted in a lesser real contact area of the CIIR rubber. This low real contact area might decrease the adhesive force. Fig. 3 shows the residual rate of the real contact area and the adhesion force of CIIR specimens treated for 5 min with oxygen plasma, as a function of the microwave power from 100 to 300 W. This figure displays the decreasing trends of real contact area while the adhesion forces remain the same. With increasing microwave power, real contact area and adhesion force decreased. This different microwave power will change the plasma density and temperature which is affected to decrease the real contact area during plasma process. Fig. 4 displays the ratio of real contact area and adhesion force by argon or oxygen gas plasma treatment with varying time. The red circles in Fig. 4 imply the ratio of real contact which is shown in Fig. 2. At 1 minute treatment time, oxygen gas was more effective than argon gas plasma treatment in decreasing the adhesion force and real contact area. However, argon plasma treatment showed a slight decrease from 5 to 10 minutes. After 15 minutes, both the real contact area and adhesion force of CIIR rubber treated with both gases decreased to almost 0%. In conclusion, oxygen and argon plasma treatments resulted in a noticeable decrease in adhesion force and real contact area.

The effect of temperature on the adhesion force between stainless steel ball and CIIR rubber is shown in Fig. 5. With 10 min treatment with oxygen and argon plasma at 200 W state, the temperature rose up to about 152.3 degree. It was maintained until 20 min treatment.

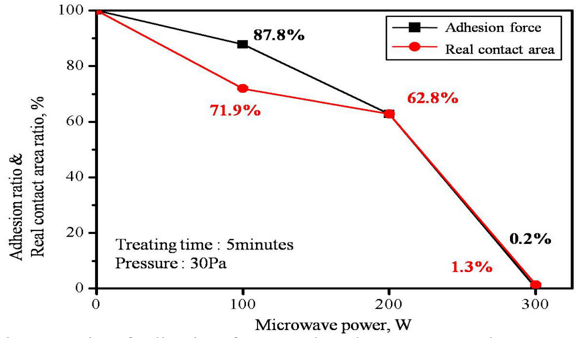

Fig. 3 Ratio of adhesion force and real contact area between stainless and CIIR after oxygen plasma treatment, as a function of the microwave power for the treatment

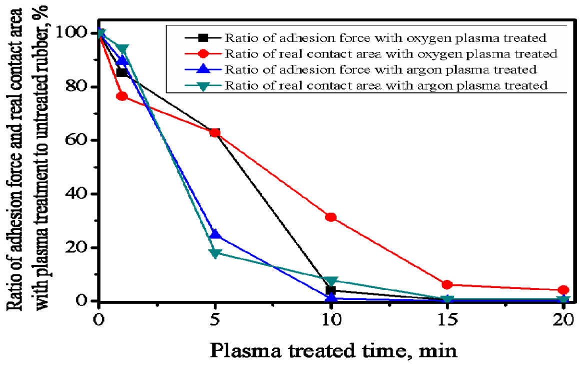

Fig.4 Ratio of adhesion force and real contact area between stainless and CIIR treated by oxygen and argon plasmas, as a function of the plasma treatment time

However, at 300 W, the temperature was higher in the oxygen plasma treated CIIR rubber than that treated with argon gas. The temperature values of oxygen and argon plasma treatments are listed in Table 1. Fig. 6 shows the adhesion forces between CIIR rubber and stainless steel ball as a function of temperature after heating with a heater and an electric furnace. In case of untreated CIIR rubber, the adhesion force was about 0.10 N as shown in the figure. For heat-treated rubber, the adhesion forces increased from 0.14 N to 0.29 N. The measured adhesion force values were not stable until 120 degree, and the adhesion forces of CIIR rubbers were 0.29 N, 0.19 N, 0.22 N and 0.24 N for 34, 39, 109.5 and 116 degree. After 120 degree, the adhesion forces decreased gradually to 0.17 N, 0.15 N and 0.14 N for 150, 165 and 215 degree. On the other hand, electric furnace showed a different trend in adhesion force of CIIR rubber as compared to that of the heater. The adhesion values were stable until 120 degree for 0.1 N. However, the adhesion forces increased after 120 degree. We believe this high variation was due to variations in the rate of reactions with oxygen and humidity in the air above the samples during the time between the heat treatment and adhesion force measurement. To summarize, Fig. 6 shows that the adhesion forces increased in all experiments after heater and electric furnace treatments. It is obvious from the results of this study that temperature has no effect on the

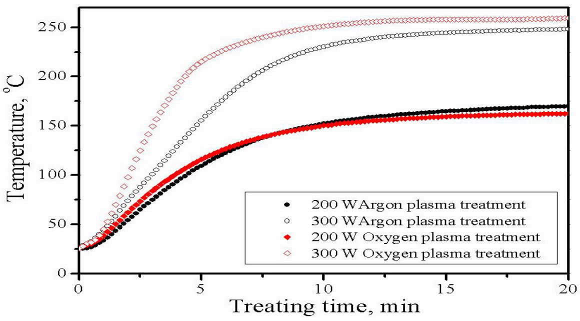

Fig. 5 Temperature variation of oxygen and argon plasma measured under 200 W and 300 W conditions

Table. 1 Temperature values of oxygen and argon plasma treatment at 200 W and 300 W conditions

(Unit: degree)

Watts	200 W					300 W
Time	1	5	10	15	20	5
Argon	34	109.5	152.3	164.8	170.1	150.0
Oxygen	39	116.0	150.3	159.4	162.7	215.1

adhesion force and real contact area of oxygen and argon plasma treated CIIR rubber. Therefore, it can be said that temperature did not affect the decrease in the adhesion force and real contact area during oxygen and argon plasma treatment. Fig. 7 shows the cross-sectional FE-SEM micrographs of untreated and oxygen plasma treated CIIR rubber. Without oxygen plasma treatment (Fig. 7a), the subsurface of the CIIR rubber appeared less granular and generally has a smoother shape. Similarly, with oxygen plasma treatment at 200 W for 5 min (Fig. 7b), visible changes in the surface were not apparent. However the subsurface of CIIR rubber pattern changed when compared to untreated CIIR rubber. And the subsurface of the CIIR rubber grew rougher with increasing treatment time (Fig. 7c). The microwave power also affected the subsurface of CIIR rubber. In this study, the 300 W 5 min treatment condition resulted in the roughest surface observed as shown in Fig. 7d in this work. This etching effect was related to the choice of plasma treatment conditions that gave different roughness in the surface morphology[4]. Fig. 7 indicates that the plasma treatment made the CIIR rubber rough according to increasing time and microwave. Also, it can be say that that the real contact area would decrease with plasma treatment in this study.

4. Conclusion

The objective of this study was find out the relationship between the real contact area and adhesion force of plasma-treated to CIIR rubber against a stainless-steel. Plasma treatments were conducted in a vacuum chamber with microwave excited high-density plasma employing oxygen and argon gases at 30 Pa. The results were as follows.

(1) At a microwave power of 200 W, the effect of treatment time on the decrease of adhesion force and real contact area was estimated; 10 and 15 min were required in argon and oxygen plasma treatments, respectively, to decrease the adhesion force by 1% compared to untreated CIIR.

(2) The residual rates of the adhesion force and real contact of CIIR showed almost similar tendency with increasing treatment time and microwave power. Therefore, it was considered that the adhesion force is strongly subjected to the real contact area.

(3) As a result of heat treatment of CIIR specimens, the adhesion forces increased from 0.1 N of untreated CIIR at all temperature levels from 34 to 215 degree. These results indicated that temperature increase during plasma treatment has no significant effect in decreasing the adhesion force.

(4) The rougher surface structure was observed with longer treatment times and larger microwave power.

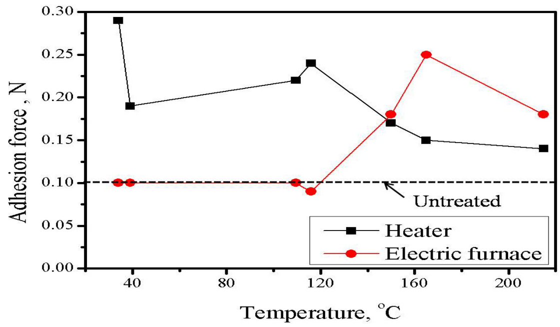

Fig. 6 The adhesion force between stainless steel ball and CIIR after heater and electric furnace treated in different temperature

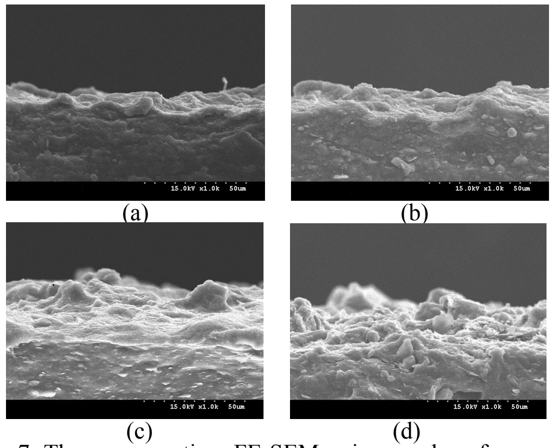

Fig. 7 The cross-section FE-SEM micrographs of oxygen plasma treated to CIIR rubber for (a) Untreated (b) 200 W, 5 min (c) 200 W, 10 min and (d) 300 W 5 min

5. Reference

[1] Kim, J. H., Kousaka, H., Umehara, N., Shimada, M. and Hasegawa, M., "Study of the Decrease in Adhesion Force between Chloride-isobutene Rubber and Stainless-Steel Ball by Plasma Treated on Rubber Surface," Advanced Materials Research, 47, 2008, 459-462.

[2] Kamiya, T., Umehara, N. and Nakamura, K., "Clarification of Governing Factors on the

Adhesion Property between Rubber and Steel and Technical Suggestion to Reduce the Adherence," Trans. of JSME. C, 74, 2008, 192-197.

[3] Nitta, I., "Visualization Technologies of Real Contact Area," Journal of Japanese Society of Tribologists, 12, 2007, 849.

[4] Grythe, K. F. and Hansen, F. K., "Surface Modification of EPDM Rubber by Plasma Treatment," Langmuir, 22, 2006, 6109-6124.