Effects of Electron Beam Irradiation on Adhesive Force of Laminated Sheet of High Strength Polytetrafluoroethylene (PTFE) and Bio-Adaptable Polydimethylsiloxane (PDMS)

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The effects of homogeneous low voltage electron beam irradiation (HLEBI) on the adhesive force of peeling (Fp) and peeling resistance energy (Ep) at each peeling probability (Pp) of laminated sheets of bio-adaptable polydimethylsiloxane (PDMS) with transparency and high strength polytetrafluoroethylene (PTFE) were investigated without glue. Although both Fp and Ep of peeling at low Pp of 0.06 were 0.2 N·m−1 and 4.0 × 10−2 J·m−1 before treatment, HLEBI enhanced the Fp and Ep up to the largest values of 11 N·m−1 and 2.2 J·m−1 of the laminated sheets irradiated at 0.13 MGy, respectively. They were more than 55 times larger than those before treatment. On the other hand, additional HLEBI reduced the Fp and Ep of laminated sheets irradiated at more than 0.22 to 0.86 MGy, although they were apparently larger than those before treatment. In order to investigate the influence of EB irradiation on Fp and Ep, electron spin resonance (ESR) signals related to dangling bonds were observed. When HLEBI cut the chemical bonds and generated dangling bonds with nonbonding electrons in PTFE and PDMS, the electrons induced the chemical bonding and intermolecular attractive force. HLEBI induced strong adhesive force of laminated sheets was explained by the discussion. Therefore, it was concluded that HLEBI was a useful tool for quick lamination of bio-adaptable PDMS and high strength PTFE. [doi:10.2320/matertrans.M2012124]

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1. Introduction

Polytetrafluoroethylene (PTFE) exhibits high wear resistance as well as high strength and fracture toughness. It can be applied to artificial blood vessel.1) When the puncture occurs at PTFE artificial blood vessel, it is difficult to restore itself. The weak puncture resistivity of plastic PTFE is a serious problem to prepare the artificial blood vessel. In order to improve the puncture resistivity, it has been expected that the PTFE is covered with soft Polydimethylsiloxane (PDMS), which exhibits high transparency and bio-adaptable and has been mainly applied to contact lenze.2) In addition, since the PDMS also shows self-adhesive,3) it can be expected to apply to wrapping the bio-medical sensors.

However, it is difficult to glue without problems induced by quick-drying glue and heat adhesion. To solve the problem, the development of rapid and safe adhesion method between PTFE to PDMS sheets has been expected to allow many biomedical applications.

Composite polymers have been prepared for biomedical applications by laminating them with heating and glue.4,5) Heating often degrades the mechanical strength and chemical properties.6) On the other hand, the glue generally volatilizes, thereby affecting human health.6) In addition, Development of rapid adhesion without glue should solve this problems.

Homogeneous low voltage electron beam irradiation (HLEBI) improves the mist resistance and wetting of inorganic materials.7) The improvement is mainly caused by the irradiation with the formation of dangling bonds in polymers.10) Dangling bonds enhance surface energy, which is probably a tool for joining the different polymers.11) Thus, rapid and safe adhesion between different polymers by using HLEBI can be expected.

In addition, the treatment time of HLEBI-sterilization is a few seconds, although sterilizing with ultraviolet light irradiation requires a few hours.12,13) Thus, HLEBI is expected to be an excellent tool for not only gluing different polymers without volatilization, but also sterilizing them for biomedical applications, simultaneously. Therefore, the effects of HLEBI on the adhesive force and its energy of peeling resistance of different polymers of PDMS film covered with PTFE have been investigated.

2. Experiment Procedure

2.1 Homogeneous irradiation of electron beam

Figure 1 shows constitutional formula of PDMS (a) and PTFE (b).

![Fig. 1 Constitutional formula of PDMS (a) and PTFE (b).](image-url)
Both stainless steel springs and 0.015 mm thick nylon6 supporting film, which loaded the compressive stress (0.080 MPa) at interface, reproducibly connected each polymer surface. As shown in Fig. 2, samples were set on the stainless spring and urethane sheets. They were compressed under 0.080 MPa by nylon6 film with 15 μm. When the compressive stress of more than 0.080 MPa was loaded for more than 1.0 h, a high reproducibility of peeling force was obtained.

Since no peeling force was observed at the interface between the nylon6 supporting film and the jig in the composites, it was easy to remove the supporting film after irradiation. The sample at the outer surface of the nylon film was homogeneously irradiated by an electron-curtain processor (Type CB175/15/180L, Energy Science Inc., Woburn, MA, Iwasaki Electric Group Co., Ltd., Tokyo). The samples were homogeneously irradiated with an electron beam through a titanium window attached to a 24 cm-diameter vacuum chamber. A tungsten filament in a vacuum was used to generate the electron beam with an electric voltage of 0.17 MeV and an irradiating current of 2.0 mA. To prevent oxidation, the samples were kept in a nitrogen atmosphere of 0.10 MPa with a residual oxygen concentration of less than 0.040%. The flow rate of the nitrogen gas was 1.5 L/s.

2.2 T-peeling test

Composite samples were prepared for the T-peeling test to evaluate the influence of HLEBI on the adhesive force ($F_p$) and its energy ($\alpha F_p$) of peeling resistance. The peeling force and its peeling distance were obtained by the peeling test (see Fig. 3), which was performed by using an micro load tensile tester (F-S Master-1K-2N, IMADA Co. Ltd., Japan). The samples were set on the stainless spring and the jig in the composites, as shown in Fig. 2. The peeling force ($F_p$) is determined by using micro load tensile tester (F-S Master-1K-2N, IMADA Co. Ltd., Japan). The adhesive force of peeling resistance ($\alpha F_p$) is estimated by the peeling load and experimental peeling width of 10 mm. The initial distance before peeling ($d_i$) is defined at the start point of peeling force, which corresponds to the start point of the first relaxation. The $d_i$ value is approximately about one mm.

(3) $F_p$ was an integrated $F_p$ as a function of peeling distance ($d_p$).

To compare to the HLEBI adhesion, the PTFE-PDMS sheets laminated by two typical commercial glues of Aronalpha (Toagosei Co., Ltd., Tokyo) and Cemedine C (Cemedine Co., Ltd., Tokyo) were prepared.

2.3 Electron spin resonance (ESR) measurement

The density of the dangling bonds was measured by means of electron spin resonance (ESR) spectrometer (JES-FA200, JEOL Ltd., Tokyo) to obtain more precise information on atomic-scale structural changes in the polymers. The microwave frequency used in the ESR analysis was in the X-band at 9.45 ± 0.05 GHz with a field modulation of 0.10 MHz. The microwave power was 1.0 mW. The magnetic field was varied from 317.0 to 327.0 mT.

3. Results

Although the large adhesive load of peeling resistance has never been measured before irradiation, the laminated sheets constructed with polytetrafluoroethylene (PTFE) and polydimethylsiloxane (PDMS) have been prepared before and after HLEBI. HLEBI laminates the PTFE with the PDMS films. Figure 4 depicts the adhesive force of peeling resistance ($F_p$)–peeling distance ($d_p$) curves of the PTFE-PDMS laminated sheet before and after HLEBI at 0.04, 0.13 and 0.86 MGy at low and mean $P_p$ of 0.06 (a) and 0.50 (b). Increasing the $d_p$ value raises the $F_p$ value. The long distance peeling gradually reduces the $F_p$ value of laminated sheet before fracture (see Fig. 4). When the mean adhesive force of peeling resistance ($\alpha F_p$) is defined, HLEBI of 0.04 to 0.86 MGy apparently raises the $\alpha F_p$.

Figure 5 plots the relationships between mean adhesive force of peeling resistivity ($\alpha F_p$) and the peeling probability ($P_p$) of PTFE-PDMS laminated sheets before and after HLEBI at each dose. All $\alpha F_p$ values of PTFE-PDMS laminated sheets irradiated of 0.04 to 0.43 MGy always exceed the all values of untreated samples. Furthermore, the $\alpha F_p$ values of PTFE-PDMS laminated sheets irradiated of 0.04 to 0.86 MGy at each $P_p$ exceed the corresponding values of untreated samples.

In order to compare to the HLEBI adhesion, the $\alpha F_p$ values of PTFE-PDMS sheets laminated by two typical commercial glues of Aronalpa and Cemedine C have been obtained. Figure 5 also shows the $\alpha F_p$–$P_p$ relationships of the laminated
sheets before and after treatments of adhesion with two typical commercial glues. The $F_p$ values of the sheets laminated by Cemendine C correspond to those of untreated samples, whereas the $F_p$ values of the sheets laminated by Aronalpha at middle and high Pp of more than 0.3 radically exceed the corresponding values of untreated samples. Since the $F_p$ values of PTFE-PDMS sheets laminated by two typical commercial glues of Aronalpha and Cemendine C at low Pp of less than 0.3 and less than 0.45 cannot exceed the corresponding values of untreated samples, respectively, both glues are not practically adaptable to laminate the PTFE-PDMS sheets. Thus, adhesion of PTFE-PDMS laminated sheets irradiated from 0.04 to 0.43 MGy is effective.

Figure 6 depicts the changes in mean adhesive force of peeling resistance ($F_p$) at each peeling probability ($P_p$) of 0.06, 0.50 and 0.94 of PTFE-PDMS laminated sheets against dose of HLEBI. HLEBI with small dosage of 0.04 to 0.13 MGy enhances the $F_p$ at each $P_p$. The maximum $F_p$ value at low $P_p$ of 0.06 of the laminated sheet irradiated at 0.13 MGy (11 N·m⁻¹) is more than 55 times larger than that before treatment. Additional HLEBI dosage of 0.23 to 0.86 MGy reduce the $F_p$ at each $P_p$, although the $F_p$ value at each $P_p$ of the laminated sheet irradiated of 0.30 to 0.86 MGy is always higher than that before treatment. Based on results of Figs. 6 and 7, HLEBI induced adhesion is effective to laminate the PTFE-PDMS sheets.
4. Discussion

4.1 Effects of two stage process on $F_s$ value

In order to estimate the adhesive force at extremely low $P_p$ value, precisely, the lowest adhesive force ($F_s$) for safety design is assumed to get the adaptable relationship of the three parameter Weibull equation with the high correlation coefficient ($f$). The peeling probability ($P_p$) depends on the risk of rupture ($\frac{\alpha F_p}{F_s}$).

$$P_p = 1 - \exp\left(-\left(\frac{\alpha F_p}{F_s}\right)^m\right)$$

In predicting the required adhesive force ($\alpha F_p$) of the new structural materials, coefficient ($m$) and constant ($F_{III}$) are the key parameters. The $F_{III}$ value is the $\alpha F_p$ value at a $P_p$ of 0.632, when the term $\ln[-\ln(1 - P_p)]$ is zero.

When $P_p$ is equal to zero, the $\alpha F_p$ value is defined as the lowest adhesive force ($F_s$). Figure 8 plots changes in the correlation coefficient ($f$) with respect to the potential $\alpha F_s$ value ($\alpha F_s$) estimated from eq. (1). The $\alpha F_s$ value is the $F_s$ value at the maximum $f$ value.

Figure 9 illustrates the linear relationships for PTFE-PDMS laminated sheets irradiated at each dosage. The values of $F_{III}$ and $m$ are determined by the least-squares method. The $m$ value is estimated by the slope of the relationship when the $F_s$ value is first decided.

Figure 10 also presents variations in the $F_s$ value of PTFE-PDMS laminated sheets against the HLEBI dose. Here, $F_s$ is always lower than the experimental $\alpha F_p$ value. The HLEBI at 0.13 MGy apparently enhances the $F_s$ of 4.8 N·m$^{-1}$, as well as the lowest experimental $\alpha F_p$ of 5.3 N·m$^{-1}$ at low $P_p$ of 0.06. Consequently, HLEBI also enhances the safety level (reliability) of PTFE-PDMS laminated sheets.
In order to utilize for practical articles, the adhesive force \( (F_p) \) has been generally required to be 50 N·m\(^{-1}\). On the other hand, 0.13 MGy-irradiated PTFE-PDMS exhibits 4.9 N·m\(^{-1}\) for the \( F_p \), 5.4 N·m\(^{-1}\) for the \( F_p \) at low \( P_p \) of 0.06 and 11 N·m\(^{-1}\) for the large \( F_p \) at high \( P_p \) of 0.94, although \( F_p \) of glued PTFE-PDMS is less than 1 N·m\(^{-1}\). HLEBI induced adhesion can be applied to practical articles with sterilization without volatilization, when the adhesive force of peeling resistivity is less than 4.9 N·m\(^{-1}\).

4.2 Enhancement of peeling resistivity by HLEBI

A peeling resistivity is defined as the slope \((dF_p/\ddp)\) of relationship between the adhesive force of peeling \((F_p)\) in Fig. 4 and peeling distance \((\ddp)\). Figure 11 plots changes in the peeling resistivity \((dF_p/\ddp)\) of PTFE-PDMS laminated sheets at each dose of HLEBI against peeling distance \((\ddp)\). The peeling gradually reduces the \( dF_p/\ddp \) of 0.06 and 0.50 of PTFE-PDMS laminated sheets without EB irradiation. HLEBI up to 0.04 MGy raises the \( dF_p/\ddp \) at small peeling distance of 0.01 mm. In addition, HLEBI enhances the \( dF_p/\ddp \) at each peeling distance of less than 0.60 mm.

The initial value of \( (dF_p/\ddp)^o\) is the maximum value of the \( dF_p/\ddp \) and is defined as the static peeling resistivity. When the fracture occurs at laminate interface, its physical meaning is stiffness just before peeling. Figure 12 plots changes in the peeling resistivity \((dF_p/\ddp)^o\) of PTFE-PDMS laminated sheets against dose of HLEBI. HLEBI up to 0.04 MGy raises the \( d(dF_p/\ddp)^o\) of PTFE-PDMS laminated sheet, whereas additional HLEBI from more than 0.22 to 0.86 MGy reduces it.

4.3 Influence of dangling bond formation induced by irradiation on mechanical properties

Figure 13 depicts the ESR signals of Polytetrafluoroethylene (PTFE), Polydimethylsiloxane (PDMS) and PTFE-PDMS laminated samples with and without irradiation at each dose. Although ESR signals could not be detected in the untreated sample, ESR signals, indicating dangling bonds formation, have been observed in PTFE, PDMS and PTFE-PDMS samples irradiated. Dangling bonds increases the free volume,\(^{18}\) which is similar to vacancy in crystal. Since HLEBI up to 0.86 MGy enhances the intensity of the ESR signals in the PTFE, it gradually enhances the density of dangling bonds.

HLEBI up to 0.04 MGy slightly enhances the intensity in the PDMS and PTFE-PDMS, although the intensity of ESR signals is weak and its dose dependence is discontinuous. Since the glass transition temperature \((T_g)\) of PDMS is below room temperature, the recovery easily occurs, resulting in weak intensity and its discontinuous dose dependence in PDMS.

ESR signals (see Fig. 13) indicate that HLEBI enhances the density of dangling bonds in PTFE. On the other hand, ESR signals are not largely detected in the PDMS samples.
even after irradiation. Since the chemical bonds in PDMS are weaker than those of PTFE, it is easy that HLEBI generates the dangling bonds. However, since it is simultaneously easy to recover quickly to annihilate the dangling bonds, they cannot be largely detected.

Dangling bonds formation of PTFE and PDMS probably correspond to the enhancements of $^pE_p$ and $^pF_p$ induced by HLEBI from 0.04 to 0.86 MGy (see Figs. 6 and 7). Since the maximum $^pE_p$ and $^pF_p$ values are found at 0.13 MGy (see Figs. 6 and 7), the low and high densities of PDMS and PTFE are found at 0.13 MGy. If the optimum density of dangling bonds exists, the maximum values can be explained.

Dangling bonds probably generate in PDMS film irradiated even at room temperature for short time. Since the air gap has not existed between PDMS and PTFE films by vacuum treatment, the adhesion force at the laminated interface has been detected. On the other hand, since the air gap has been exists between the support film and sample, the adhesion between nylon6 support film and specimen does not occur.

As shown in Fig. 1, the PTFE is constructed with fluorine and carbon, whereas the PDMS is formed by carbon, hydrogen, silicon and oxygen. Figure 14 illustrates Fluorine (1s) signal in PDMS side (a), Si (2s) signal in PTFE side (b) and oxygen (1s) signal in PTFE side (c) of X-ray photo-electron spectrometer (XPS) (Quantum 2000, ULVAC Co., Japan) surface analysis of PTFE-PDMS laminated films with and without HLEBI. The element of fluorine is also found in the PDMS side peeled surface after HLEBI (see Fig. 14(a)). The elements of silicon and oxygen are found in the PTFE side of peeled surface (see Figs. 14(b) and 14(c)). Their concentrations of PTFE part with HLEBI are higher than those without HLEBI. Thus, the HLEBI adhesion induces mass transports to each other at interface of the PTFE-PDMS laminated sheet.

Figure 15 shows schematic illustration of bonding sites at interface between PTFE and PDMS. When HLEBI cuts the chemical bonds and generates dangling bonds with nonbonding electrons in PTFE and PDMS polymers, the electrons probably induce the chemical bonding and intermolecular attractive force. HLEBI induced strong adhesive force of laminated sheets is explained by the discussion. Therefore, it is concluded that HLEBI is a useful tool for quick lamination of bio-adaptable PDMS and high strength PTFE.

Since the dangling bonds are generated by HLEBI less than 0.13 MGy (see Fig. 13), they act as bonding sites to terminated atoms of another side by coulomb attractive force at the adhesive interface between PTFE and PDMS. Thus, the high adhesive forces of peeling resistance ($^pF_p$) (Fig. 6) and high adhesive energy of peeling resistance ($^pE_p$) (Fig. 7) can be explained.

When additional dangling bonds generated by HLEBI at more than 0.22 to 0.86 MGy act as crack origins and propagation sites at the laminated interface between PTFE and PDMS, HLEBI at more than 0.22 to 0.86 MGy degrades the polymers and then reduces the $^pF_p$ and $^pE_p$ (see Figs. 6 and 7). Furthermore, the electrons probably induce the polarization at terminated atoms of polymers at the adhesive interface. In addition, the additional dose of HLEBI enhances the density of dangling bonds. When the density is too much, the intermolecular force to attract to each other side should reduce, resulting in decay of adhesive $^pF_p$ and $^pE_p$ (see Figs. 6 and 7).
4.4 Discussion of irradiation depth in polymer

Christenhusz and Reimer\(^{[19]}\) have evaluated the depth of penetration of EB irradiation of less than 0.20 MV electrical potential by measuring the Joule heat generated in copper films.\(^{[19]}\) Based on density ($\rho$: kg m\(^{-3}\)) and potential drop ($\Delta V$: kV), penetration depth ($D_{th}$: m) is given by

$$D_{th} = 66.7\Delta V^{5/3}/\rho$$

(2)

The potential at the surface sample ($V_s$) is expressed by the following equation.\(^{[19]}\)

$$V_s = V_o - \Delta V$$

(3)

Based on the assumption of Christenhusz and Reimer,$^{[19]}$ the potential drop ($\Delta V$) is caused by the loss of irradiation energy of the electron beam from the initial potential at the filament surface of the electron beam gun ($V_o$) to the potential at the surface sample ($V_s$). The potential drop ($\Delta V$) is expressed by the following equation.\(^{[19]}\)

$$\Delta V = \tau \cdot V_o / S$$

(4)

Here, $\tau$ and $S$ are thickness of the Ti sheet and mass thickness nitrogen gas, respectively.

The electrical potential (0.13 MeV) is estimated from the electrical potential (0.17 MeV), the thickness (10 $\mu$m) of the titanium window ($\rho = 4.6$ g cm\(^{-3}\)), and the distance (30 mm) in the nitrogen gas atmosphere ($\rho = 1.1$ kg m\(^{-3}\)) between the surface of the sample and the window. Since the electrical potential is 0.13 MeV, eq. (2) predicts that the penetration depth ($D_{th}$: m), estimated by Christenhusz and Reimer,$^{[19]}$ is 0.256 mm for PDMS and 0.105 mm for PTFE, when the densities ($\rho$) are 1.0 Mg m\(^{-3}\) for PDMS and 2.1 Mg m\(^{-3}\) for PTFE, respectively.

The penetration depth Libby assumption\(^{[20]}\) of HLEBI related to mass thickness ($S$: mg cm\(^{-2}\)) and irradiation voltage ($V_s$/kV), is expressed by

$$S = 1/150 \times \Delta V^{5/3}.$$  

(5)

The estimated mass thickness is 35 mg cm\(^{-2}\), when the initial irradiation potential is 0.17 MeV. Since the mass thickness of the Ti foil (1.8 mg cm\(^{-2}\)) and N\(_2\) gas (0.15 mg cm\(^{-2}\)) reduces the penetration depth of HLEBI, the mass thickness ($S$) of the polymer sample is 33 mg cm\(^{-2}\).

In addition, based on the Libby assumption,\(^{[20]}\) the electrical potential of the sample surface is expressed by

$$\Delta V = (150 \times S)^{3/5}.$$  

(6)

Furthermore, the penetration depths of PDMS (0.378 mm) and PTFE (0.152 mm) are obtained by the assumptions of Libby.\(^{[20]}\)

The sample is positioned in an aluminum plate holder (0.15 m $\times$ 0.15 m) and is transported on a conveyor at a speed of 10 m/min. Because the minimum dose of HLEBI is 43 J/g (1.1 $\times$ 10\(^{13}\) e m\(^{-2}\) = 0.043 MGy), each burst of irradiation is performed for a short time (0.23 s) at 0.043 MGy to avoid excessive heating of the sample. As a result, the temperature of the sample surface remained below 323 K just after irradiation. Both surfaces of the samples are repeatedly irradiated to increase the total irradiation dose. The interval between the end of one period of irradiation and the start of the next is 30 s. The dosage is proportional to the yield determined from the irradiation current, the conveyor speed, and number of irradiations.

The yield is calibrated using Radio chromic nylon film (FWT-60-00: FAR WEST TECHNOLOGY Inc., Goleta, CA, USA) and dose-meters (FWT-92D: FAR WEST TECHNOLOGY Inc., Goleta, CA, USA).

The electron number density ($N_g$) is expressed by the following equation in metals.\(^{[21]}\)

$$N_g = DE/C_g$$

(7)

where, $N_g$ (e m\(^{-2}\)) is the electron number per unite volume of 1.0 m\(^2\), the irradiation dose (cm\(^{-2}\)), $DE$ (Sv s\(^{-1}\)) is the dose equivalent per unite second (Sv s\(^{-1}\)), and $C_g$ (Sv s\(^{-1}\))(e m\(^{-2}\)) is the transferred coefficient of particle flux and dose equivalent \(0.17 \times 10^{-13}\) (Sv s\(^{-1}\))(m\(^2\)e m\(^{-2}\))\(^{-1}\).\(^{[21]}\)

Since the irradiated depth of PDMS of supporting film (75 $\mu$m thickness) and adhesive film (30 $\mu$m thickness) in the composites is 0.11 mm, the adhesive interface is perfectly irradiated. Based on the gotten values in Table 1, the whole samples are homogeneously irradiated, because the effective depth is 0.317 $\pm$ 0.061 mm.

5. Conclusion

The effects of homogeneous low voltage electron beam irradiation (HLEBI) without glue but with sterilization on the adhesive force of peeling ($E_{fi}$) and its energy ($E_p$) of laminated sheets constructed with polydimethylsiloxane (PDMS) and polytetrafluoroethylene (PTFE) were investigated.

(1) Although $E_{fi}$ and $E_p$ at low peeling probability ($P_{up}$) of 0.06 were 0.2 N m\(^{-1}\) and 0.04 J m\(^{-1}\) before treatment, HLEBI enhanced the $E_{fi}$ and $E_p$ up to the maximum values of 5.3 N m\(^{-1}\) and 1.1 J m\(^{-1}\) of the laminated sheets irradiated at 0.13 MGy, respectively. They were more than 55 times larger than those before treatment.

(2) On the other hand, additional HLEBI reduced the $E_{fi}$ and $E_p$ of laminated sheets irradiated at more than 0.22 to 0.86 MGy, although they were apparently larger than those before treatment.

(3) In order to investigate the influence of EB irradiation on $E_{fi}$ and $E_p$, electron spin resonance (ESR) signals related to dangling bonds were observed. When HLEBI cut the chemical bonds and generated dangling bonds with nonbonding electrons in PTFE and PDMS, the effects of electron beam irradiation on adhesive force of laminated sheet of high strength (PTFE) and bio-adaptable (PDMS) 1663
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