ANALYSIS OF CONDUCTIVITY DISTRIBUTION IN PI IRRADIATED BY PROTON USING SIMULTANEOUS MEASUREMENT OF SPACE CHARGE DISTRIBUTION AND CONDUCTION CURRENT

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ABSTRACT

We prepared 2 type of commercially available polyimide (PI) which is irradiated by a proton, we evaluate the conductivity distribution of those PI using simultaneous measurement of space charge distribution and conduction current through a bulk. As the result, we obtained difference tendency of conductivity in each PI. In PI-1, no significant conductivity distribution was not observed in the bulk. On the other hand, in PI-2, we found that conductivity is distributed in the bulk. The conductivity value at the position of the maximum penetration depth is 10 times larger than other region of bulk.

1. INTRODUCTION

Polyimide (PI) and fluoride materials are used for multi-layer insulator and optical solar reflector, respectively. Those materials are for keeping constant temperature of satellite inside. Those materials are always exposed high energy charged particles, such as electrons and protons, on orbit. Electrons and protons are injected into the bulk of those insulators, and accumulated in the bulk of them. Concerning the study for charge accumulation in the polymeric materials irradiated by an electron, our group and Griseri et al[1][2]. have carried out. However, concerning the study for charge accumulation in the polymeric materials irradiated by a proton, any researcher did not carried out. Therefore, our research group try to understand the characteristic of charge accumulation and dielectric properties in PI and fluoride materials irradiated by a proton.

When those insulating materials are irradiated by charged particles, it is considered that the degradations of electrical properties are produced due to scission of molecular chains and generation of the defects. Actually, when the DC high electric filed was applied to the proton irradiated PI, space charge polarization by generated electron-hole pairs were produced in the bulk. As those electron-hole pairs enhanced the electric filed in bulk, it is considered that the possibility of breakdown is increased.

Therefore, we need to investigate the characteristic of electrical property, such as conductivity and space charge accumulation in the bulk. From our previous research work, we measured space charge distribution in the bulk of PI during proton beam irradiation using pulsed electroacoustic (PEA) method [3][4]. As the results, positive charge amount reached maximum immediately after irradiation, and gradually decreased during irradiation. It is considered that this phenomenon was produced due to radiation induced conductivity (RIC) stimulated in the region of proton penetration [5].

In this report, in order to confirm the effect of RIC in the proton irradiation area, we prepared 2 types of commercially available PI, we evaluated the conductivity distribution of those irradiated PI using the results of simultaneous measurement system between conduction current and space charge distribution under DC stress.

2. MEASUREMRNT SYSTEM

2.1. Principle of pulsed electro-acoustic method

The schematic diagram of the pulsed electro-acoustic (PEA) method is shown in Fig. 1[3][4]. A sample is sandwiched between high voltage and grounded electrode. When the pulsed voltage is applied to the sample though those electrodes, the pulsed pressure waves are produced from each charge accumulated position by the coulomb's force. Those pressure waves are propagated thought the bulk and electrode, and are detected at the piezoelectric device. The intensity and time distance of detected signal is proportional to the amount of accumulated charges and position of each accumulated charges.

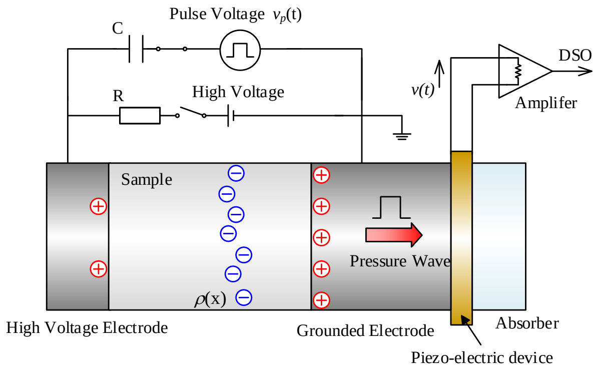


Figure 1. Schematic view of PEA measurement method

2.2. Simultaneous measurement of space charge distribution and conduction current[6]

The schematic diagram of simultaneous measurement of space charge distribution and conduction current is shown in Fig. 2. Measurement electrode disposed on the lower side of the sample is isolated by PTFE. Therefore, it is possible to measure the conduction current which is passed through the bulk by suppressing the influence of the leakage current from the sample surface. Further, the bottom of the measurement electrode is installed quartz glass. The pressure waves are able to propagate through the glass, and detected at the piezoelectric device. Moreover, a coaxial switch which is inserted between a measurement electrode and an ammeter meter make it possible to measure the space charge distribution and the external current in the same sample.

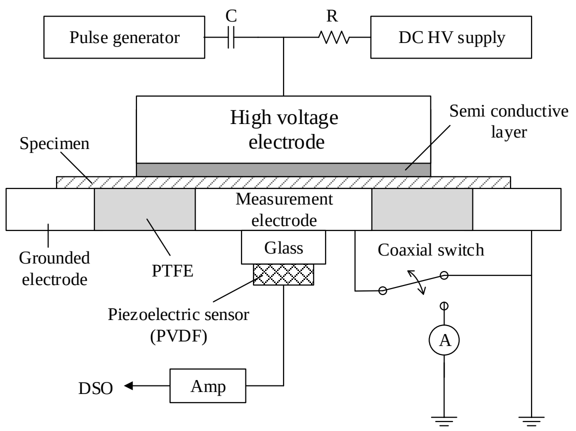


Figure 2. Schematic view of space charge distribution and conduction current simultaneous measurement system

2.3. The calculation of the conductivity distribution

It is possible to perform a space charge distribution and current measurement at the same time in this measurement system. The displacement current distribution $J_d(x,t)$ is possible to calculate from the space charge distribution $\rho(x,t)$ and electric field distribution $E(x,t)$. The external current density $J_e(x,t)$ is expressed as the equation (1) which is using the conduction current density $J_c(x,t)$ and the displacement current density $J_d(x,t)$.

$$J_e = J_c(x,t) + J_d(x,t) \tag{1}$$

The displacement current density is expressed as the time displacement of the electric field as shown in the equation (2).

$$J_d(x,t) = \varepsilon \frac{\partial E(x,t)}{\partial t} \tag{2}$$

The conductivity distribution is expressed as the equation (3) which is using the conduction current density and the electric field distribution.

$$J_c(x,t) = \kappa(x,t)E(x,t) \tag{2}$$

Therefore, conductivity distribution $\kappa(x,t)$ can be calculated from the result of simultaneous measurement of the space charge distribution $\rho(x,t)$ and the external current density $J_e(x,t)$.

3. MEASUREMRNT PROTOCOL

The samples are 2 types of commercially available PI film, called PI-1, PI-2, respectively.

The experiment of proton beam irradiation was carried out in vacuum atmosphere of approximately 1×10^{-5} Pa. Concerning the irradiation condition, the proton was irradiated with energy of 2.0 MeV and 30 nA/cm^2 for 30 minutes. The irradiation was carried out using 3 MeV tandem type ion accelerator facility of Takasaki Advanced Radiation Research Institute of Japan Atomic Energy Agency (JAEA). After irradiation, we measured space charge distribution and the conduction current in irradiated sample under DC average stress of 100 kV/mm. The experiment was started after 1 and 3 days.

4. RESULT AND DISCUSSION

4.1. Result of the PI-1

Simultaneous measurement results of space charge distribution and conduction current is shown Fig. 3. In the Fig, (A)-(C) shows the result of non-irradiated sample, 1 day relaxation sample and 3 days relaxation sample, respectively. Furthermore, (a)-(d) also shows the external current density, space charge distribution, electric field distribution and conductivity distribution with voltage application time progress, respectively. In Fig. 3(a), the vertical and the horizontal axes show the external current density J_e [nA/m^2] and the time t [min], respectively. The black and red line shows result of non-irradiated and irradiated sample of external current density in Fig (a). In Fig. 3(b)-(d), the vertical and the horizontal axes show the position x [μm] and the time t [min], respectively. The magnitude of the quantity of each measurement results are described using colors and values of the colors are corresponding to the scale of color bars. The broken horizontal line in the bulk of figure (b)-(d) shows the proton penetration depth calculated by "SRIM" as a numerical simulation method [7].

(A) Non-irradiated sample

From Fig. 3(A)-(a), external current density was increased to the maximum value immediately after applying high voltage, and it has been observed to decay to approximately 1.7×10^2 nA/m^2 until the end of measurement. From Fig. 3(A)-(b), it was observed that the accumulation of same polarity charge in the vicinity of each electrode. It is considered that same polarity charges are injected from the each electrode by applied electric field. In addition, from Fig. 3(A)-(d), we observed that conductivity distribution was not change.

(B) 1 day relaxation sample from irradiation

From Fig. 3(A)-(a), the external current density was increased to the maximum value immediate after DC application with the same as non-irradiated sample, and it has been observed to decay to approximately 1.4×10^3 nA/m^2 until the end of measurement. The value of current density becomes about 10 times larger than non-irradiated sample. From Fig. 3(B)-(b), the accumulated positive charge at the vicinity of the proton penetration depth and cathode was observed, and accumulation of negative charge in the vicinity of anode was observed concerning these charge accumulation phenomena is called the space charge polarization. From Fig. 3(B)-(c), the electric field was enhanced 130 kV/mm due to charge accumulation in the bulk. From Fig. 3(B)-(d), although the external circuit current is increased 10 times larger than non-irradiated sample's current, the

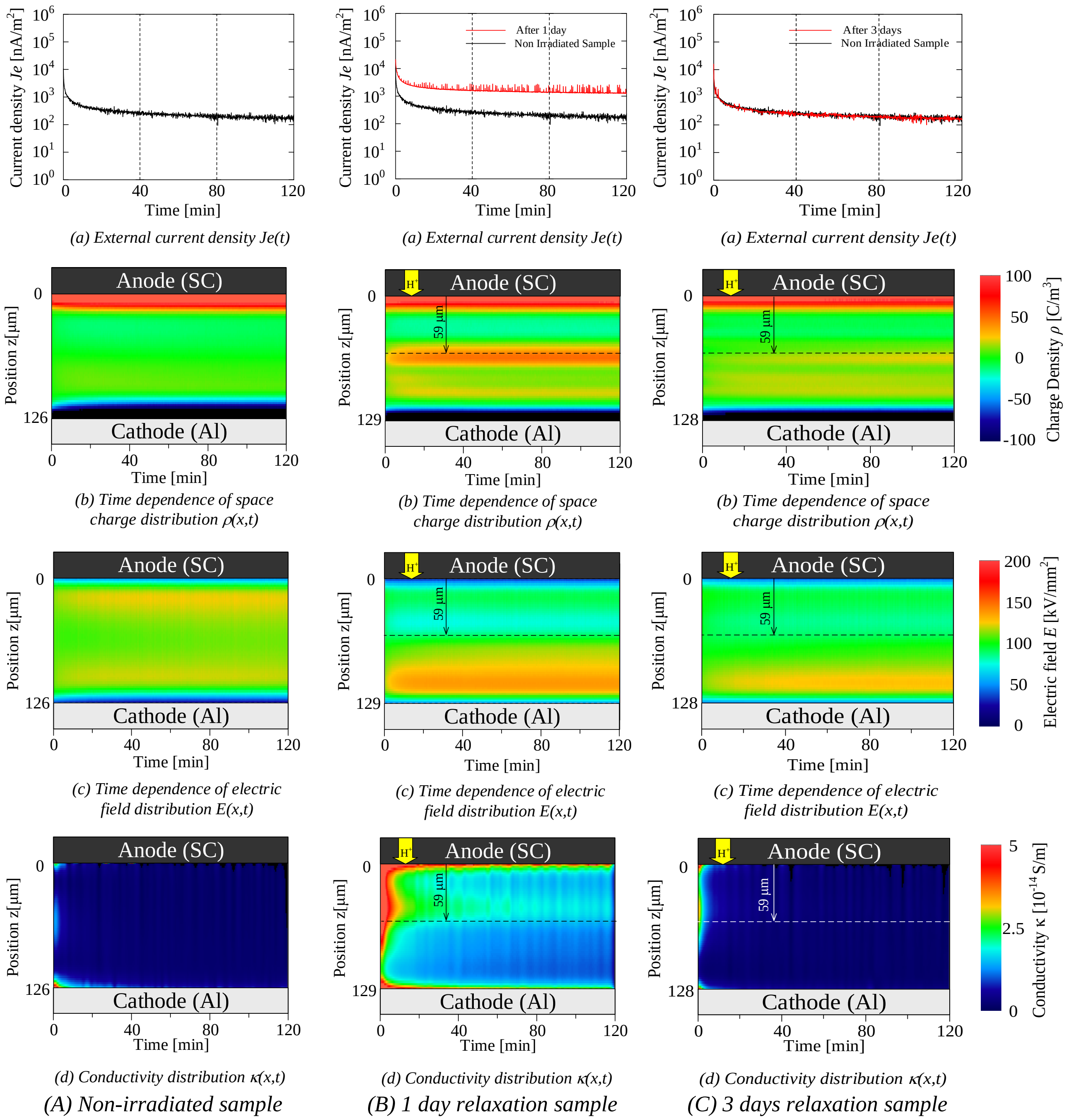


Figure 3. Measurement results of non-irradiated and irradiated PI-1 under DC stress

conductivity overall was increased inside PI-1.Moreover, the conductivity was increased more than 5 times in the vicinity of proton penetration depth Immediate after DC application.

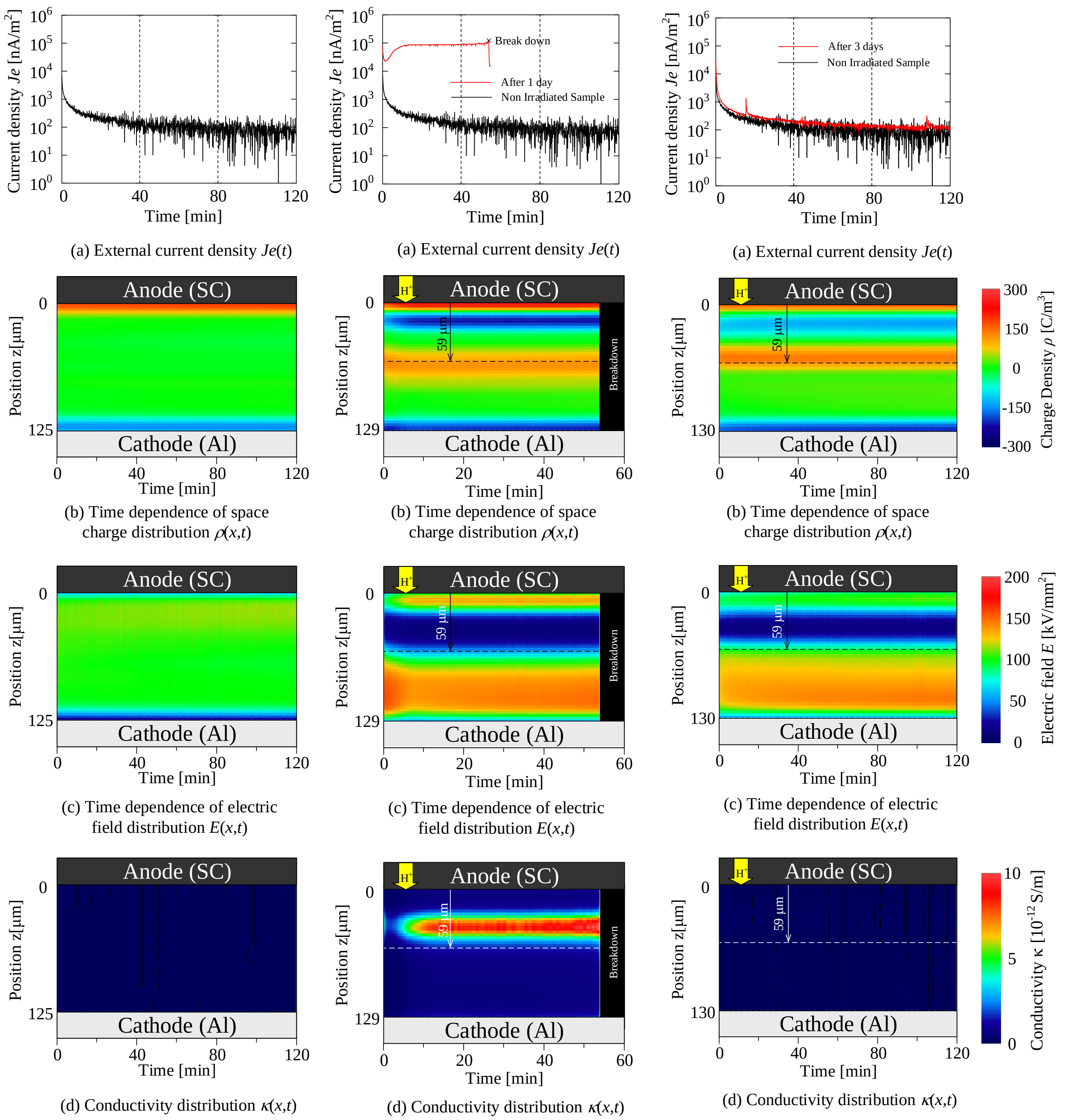
(C) 3 days relaxation sample from irradiation

From Fig. 3(C)-(a), the value of the external current density indicates a nearly equivalent to the non-irradiated sample. From Fig. 3(C)-(b), space charge polarization was also observed in the bulk with the same as 1 day relaxation sample from irradiation. Moreover, from the comparison with the measurement result of 1 day relaxation sample, it is possible to be seen that the space charge accumulation amount is reduced. From Fig. 3(C)-(c), the electric field was enhanced up to about 125 kV/mm.

4.2. Result of the PI-2

(A) Non-irradiated sample

The measurement result of PI-2 is shown Fig. 4. From Fig. 4(A)-(a), external current density was increased to the maximum value immediate after DC application, it has been observed to decay to approximately 1.4×10^2



(A) Non-irradiated sample *(B) 1 day relaxation* *(C) 3 days relaxation*

Figure 4. Measurement results of non-irradiated and irradiated PI-2 under DC stress

nA/m^2 over time. From Fig. 4(A)-(b), the space charge accumulation was not observed in the bulk of the non-irradiated sample. From Fig. 4(A)-(d), the conductivity was uniform value.

(B) 1 day relaxation sample from irradiation

From Fig. 4(B)-(a), external current density was decayed to approximately 2.3×10^4 nA/m^2 after DC application, subsequent current density increased to approximately 1.0×10^5 nA/m^2 until the end of measurement. At this time, PI-2 was led to the breakdown. Moreover, the external current density of irradiated PI-2 increased 10^3 times larger than the non-irradiated sample. From Fig. 4(B)-(b), space charge polarization was also observed in the bulk. Conspicuous changes in the space charge behavior inside the bulk was not observed over time. From Fig. 4(B)-(c), the electric field enhancements increase inside the bulk, and reach to about 150 kV/mm due to the space charge polarization. From Fig. 4(B)-(d), conductivity was locally increased at the vicinity of the proton penetration depth, we also confirmed that the increase of conductivity was from 4.0×10^{-13} S/m to 1.1×10^{-12} S/m. In particular, the conductivity in the vicinity of proton penetration depth was increased about 10 times larger than other regions of the bulk.

(C) 3 days relaxation sample from irradiation

From Fig. 4(C)-(a), external current density was increased to the maximum value immediate after DC application with the same as non-irradiated sample, it has been observed to decay to approximately 1.1×10^2 nA/m^2 until the end of measurement. However, space charge polarization was still observed. From Fig. 4(C)-(c), the electric field was enhanced up to about 150 kV/mm due to space charge polarization.

4.3. Discussion

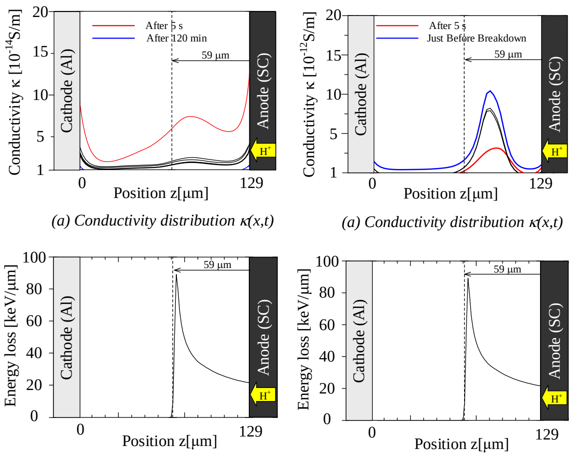
From the above results, it is consider that the origin was produced due to radiation induced conductivity (RIC) by the proton irradiation, and conductivity of the proton passage region is increased. The main factors of RIC are activation of the sample, scission of molecular chains, the occurrence of ionization in the material, and linear energy transfer (LET) of the irradiated particles to the bulk of material.

In this measurement result, space charge polarization in the irradiated sample after DC application was observed. It is considered that the electron-hole pairs were generated due to scission of molecular chains by proton irradiation, and polarized by the high voltage application. From the comparison PI-1 and PI-2 of space charge distribution, it is considered that charge injection from the electrode was produced due to reduction of injection barrier for holes, or it is considered that electron sweeping to the anode. Furthermore generated electron-hole pairs are considered to rapidly recombined in PI-1 compared with PI-2. Therefore, by performing the calculation of the energy level caused by or molecular orbital method for performing a molecular structure analysis of proton irradiation sample, it is necessary to consider recombination probability of electron-hole pairs.

Next, it describes the change over time after irradiation of the space charge distribution and external current density. From the measurement result of Fig. 2 and 3, the attenuation of current density and accumulated charge amount was observed in PI-1 and PI-2 with relaxation time progress.

However, the space charge accumulation in 3 days relaxation sample of PI-2 had been observed. It is considered that the effect of the RIC resulting from the scission of molecular chains by the incident of proton irradiation still remained after irradiation in PI-2. This phenomenon is well known as delayed radiation-induced conductivity (Delayed Radiation Induced Conductivity: DRIC) [(8)]. It is considered that conductivity was increased in PI-2, and was attenuated more quickly in PI-1. The effect of DRIC has been reported to be reduced by the relaxation time progress after the irradiation. This electron-hole pairs generated by high-energy proton irradiation is recombined with the relaxation time progress, it is considered the space charges consisted of electrons and holes produced due to irradiation are became less. From the above, the effect of DRIC is seemingly decreased with the relaxation of time progress.

Regarding conductivity of irradiation samples, it was examined LET caused by the interaction of protons and materials. Conductivity distribution waveform of 1 day relaxation sample, and the calculated result of LET by ionizing are shown in Fig. 5. In Fig. 5-(a), the vertical and the horizontal axes shows the conductivity κ [S/m] and the position x [μm], respectively. In Fig. 5-(b), the vertical and the horizontal axes shows energy loss [keV/μm] and the position x [μm], respectively. The energy loss was calculated using Transport of Ions Matter (TRIM). From Fig. 5-(a), conductivity distribution of PI-1 has the maximum value in whole of bulk, but it has been observed to decay with the relaxation time progress. However, the conductivity distribution of PI-2 has the maximum value at the in front of the maximum penetration. From Fig. 5-(b), energy loss due to ionization is occurred in proton irradiation region. Moreover, it is observed to be the maximum value in the vicinity of maximum penetration depth of proton. Therefore, it is considered that carriers were generated easily in the position which the irradiation proton lost a lot of energy, and become partial conductivity increase. It is necessary to evaluate the relationship of LET and conductivity by performing simultaneous measurement of space charge distribution and conduction current, when the acceleration energy of irradiation proton is changed.



(b) Linear Energy Transfer distribution (b) Linear Energy Transfer distribution

(A)PI-1 *(B)PI-2*

Figure 5. Liner energy transfer distribution in PI

5. CONCLUSION

We prepared 2 type of PI which is irradiated by a proton, we evaluate the conductivity distribution using simultaneous measurement of space charge distribution and conductivity current. In the space charge distribution measurement, accumulation of charge in the non-irradiated sample was not observed. However, accumulation of positive and negative charges in irradiated sample was observed. In the external current density measurement, the current density of the irradiated samples were increased compared to non-irradiated samples, confirmed the conductivity increases from this. From this result, it is considered that the electron-hole pairs are generated due to scission of molecular chains by proton irradiation. The conductivity distribution of irradiated PI-2 was locally increased in the vicinity of the proton penetration depth, and the conductivity in the vicinity of proton penetration depth was increased about 10 times, which was compared to the other regions. There is conductivity distribution in the interior of irradiated PI, and a difference in residual effect of RIC the material. Further, it is necessary to acquisition of reproducibility, and consider about conductivity distribution.

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