Secondary Electron Emission and Surface Charging Evaluation of Alumina Ceramics and Sapphire

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ABSTRACT

The breakdown of alumina rf windows is mostly caused by multipactor, as well as by material defects and contamination. Since multipator induces localized surface heating, leading to surface melting, it is necessary to observe secondary electron emission (SEE) coefficients of alumina ceramics under high temperature conditions. The SEE coefficients of commercial alumina ceramics and sapphire were measured by a scanning electron microscopy (SEM) with a single short-pulsed electron beam (100 pA, 1 ms) at room temperature and at 650°C. Additive materials used for sintering alumina, such as SiO_2 and MgO, were also investigated. Surface charging evaluations have also become important because the accumulated charges are discharged at the threshold field, resulting in surface discharge. The surface charging evaluations were carried out by multi-pulse measurements with the injection of successive pulses on the sample. As a result, reductions in the SEE coefficients with temperature were confirmed, except for sapphire. The multi-pulse measurement results indicated that surface charging of the sapphire was higher than that of other samples. This may be one of the factors that causes sapphire not to be durable for rf window applications, compared with alumina ceramics. Although there are few exceptions, it was found that the SEE coefficients of alumina ceramics increased with the purity and the average grain size.

Index Terms — **Electron emission, surface charging, high-temperature measurement, multipactor, rf window, alumina ceramic, sapphire.**

1 INTRODUCTION

ALUMINA ceramic is widely used as electrical insulators in vacuum applications, such as klystrons, because of its excellent characteristics, such as high mechanical strength, high resistivity at high temperature, small microwave loss, and less gas emission. Klystrons are used as microwave sources to accelerate charged particles in high-energy accelerators. Alumina ceramics are used as rf windows between acceleration tubes and klystrons to pass the microwaves and to seal the vacuum.

The requirements of high-power klystrons currently increase because the demand for acceleration energy increases. One of the most serious problems for developing a high-power klystron is surface flashover along the alumina rf window [1-3]. Many theories have been proposed to explain the surface flashover mechanism along insulators [4-6]. The most generally accepted mechanism involves the "secondary electron emission avalanche (SEEA)" model [6]. According to this model, flashover may be initiated by primary electrons emitted from a triple junction. These primary electrons are then accelerated and multiplied due to the high secondary electron emission coefficient of insulators. Unfortunately, this multiplication of secondary electrons (multipactor) induces excess surface heating, leading to the localized surface melting. Therefore, studies on the SEE coefficient of insulators under high temperature conditions are required to understand the surface flashover process.

Extensive studies on the secondary electron emission (SEE) have been carried out since 1902 by Austin and Starke [7], and are presently being thoroughly studied by many researchers. Many theories based on these studies have been proposed to explain the SEE phenomena [7-13]. However, there is still little experimental data concerning SEE under high temperature conditions, especially for technical materials, such as alumina ceramics. The present study was intended to measure the SEE of various kinds of

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commercial alumina ceramics for electrical insulation use in vacuum at room and high temperatures. Sapphire (single crystal alumina) and sintering additive materials, such as SiO_2 and MgO, were also investigated.

It has been revealed that the SEE of an insulator decreases with electron irradiation due to surface charging. Surface charging on an rf window can be harmful from the viewpoints of the pile-up of charges because the accumulated charges set up local electrical fields and may be discharged when the field exceeds the breakdown threshold field of the material. In order to evaluate surface charging, a multi-beam irradiation method was adopted and surface charging of the commercial alumina ceramics was also summarized.

2 EXPERIMENTAL SETUP

2.1 SEE MEASUREMENT SYSTEM

SEE measurements were carried out using a scanning electron microscopy (SEM) at room and high temperatures [14]. The sample stage of the SEM was modified to enable it to heat a sample up to 750°C. The vacuum pressure during the measurement was around 10^{-4} Pa. A schematic diagram of the measurement system is shown in Figure 1. A single pulsed primary beam of 100 pA for 1 ms duration with a quite low current density (3 $\mu C/m^2$) was adopted, since long-pulse irradiation (>10 ms) and a large number of irradiation (>10 times) can induce serious charging on the surface. The primary current was measured at a biased (+40 V) Faraday cup made of graphite. The secondary current was captured by a biased heat-reflector coated with a DLC (diamond-like-carbon) film which has low SEE coefficients to minimize the SEE from the reflector surface. SEE coefficient was defined as the ratio of the secondary to primary current at the beginning of the pulses.

It is well known that a large number of secondary electrons that are emitted from an insulator results in surface charging on an insulator. The pile-up of charges on an rf window due to multipactor is not desirable, because the accumulated charges are discharged at the breakdown field threshold. To evaluate surface charging due to secondary electron emissions, multi-pulse measurements were carried out under room and high temperatures. In these measurements, 4 or 5 successive pulses (100 pA, 1 ms) were irradiated on the insulator at a same site. Patterns A and B of successive pulses, as shown in Figure 2, were employed. Although the total irradiation charges became 4 or 5 times larger than that from the single pulsed-beam method, both SEE and surface charging could be estimated. Pattern A shows irradiation with the same intervals of 1 ms (Figure 2a). If a larger decrease of the SEE was observed with successive pulses, the material had higher surface charging. Pattern B shows irradiation of 4 successive pulses with elongated intervals (Figure 2b). The results are useful to evaluate the recovery time constant of surface charging. If the SEE at the 4th pulse was larger than the SEE at the 3rd pulse, it meant that the SEE recovered by the 4 ms interval, corresponding to a typical recovery time of 4 ms. A digital to analog converter (DAC) board, which was installed in a PC, triggered primary electron injection of both patterns. Since the multipactor takes took place at less than 5 keV injection [15], the incident energy of 1 keV was utilized for patterns A and B.

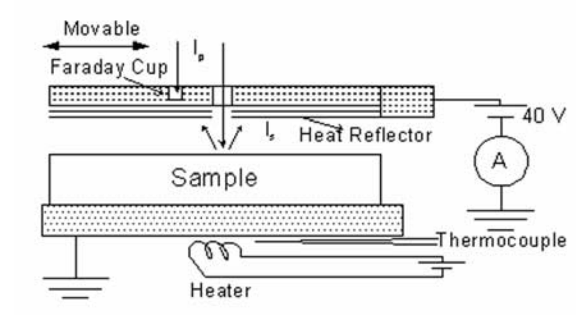

Figure 1. A schematic diagram of the SEE measurement system.

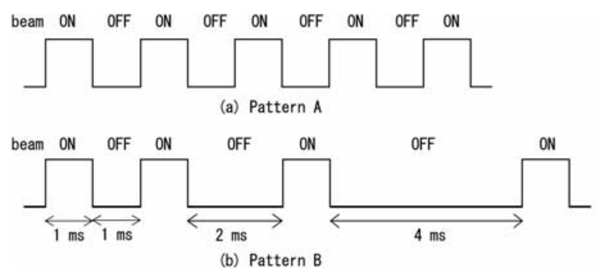

Figure 2. Two patterns of the successive pulses (100 pA, 1 ms, 1 keV) employed in multi-pulse measurements.

2.2 EXAMINED SAMPLES AND ANNEALING TREATMENT

The examined samples (19 mm in diameter) were several kinds of commercial alumina ceramic disks, which are used for electrical insulation in vacuum and single-crystal alumina (sapphire). The properties of the samples are listed in Table 1. SiO_2 and MgO, which are the common additives in the

Table 1. Properties of the examined sample.

Sample	Purity [%]	Specific gravity [g/cm^3]	Flexural strength [MPa]	Average grain size [µm]	Loss tangent	Comment
HA92	92	3.6	350	4	8.0E-04	
HA960	96	4.0	500	3	4.0E-05	crystallized grain boundaries
UHA99	99	3.9	520	4	1.0E-04	dense structure
KP990	99.5	3.9	500	4	3.0E-04	
SSA-S1	99.6	3.91	330	15	3.0E-05	
HA997	99.7	3.9	300	10	4.0E-05	rf window
KP999	99.9	3.9	320	15	9.0E-04	
Sapphire	100	3.98	---	---	3.0E-05	single crystal
MgO	100	3.6	---	---	---	single crystal
SiO_2	100	---	---	---	---	single crystal

commercial alumina ceramics, are also measured. HA997 is currently used as a high-power rf window material, and UHA99 was used before [16]. Michizono et. al. [17] reported that both alumina ceramics are more durable for rf window applications compared with sapphire. HA960 is specially sintered to make the boundary additives crystallized, which contributes to reducing the dielectric loss tangent to be as small as that of sapphire.

It is well-known that alumina ceramics may have trapped charges in their vacancies. Trapped charges are not desired, because they influence the secondary electrons emitted from the alumina ceramic. Sato et. al. [18] reported that an annealing treatment in the atmosphere at 1500°C for 1 hour is effective to remove the mechanical stress of insulators, which affects their properties concerning reduction of the trapped or stored charges in the vacancies of alumina ceramics, especially for relatively low purity alumina (about 90%). Therefore, all of samples examined in this study were annealed in the atmosphere for 1 hour before the measurements. The annealing temperatures were 600°C for SiO_2 and 1400°C for other samples.

3 RESULT AND DISCUSSION

3.1 SEE COEFFICIENTS

3.1.1 ALUMINA CERAMICS

The SEE coefficients of commercial alumina ceramics were measured at room temperature and at 650°C. Figure 3 shows the relationship between the SEE coefficient of HA92 (92% purity) and the incident energy of the primary electrons at room and high temperatures. The SEE coefficient increased with the primary energy for low energies, then went to the maximum value, and finally decreased for high primary energies. At 650°C, the SEE coefficients of the HA92 were lower than those obtained at room temperature. The decrease was not caused by any surface modifications, because the SEE coefficients after cooling down were almost the same as the previous values at room temperature. Temperature dependence on SEE coefficients of other ceramic samples was also examined (Figure 4). The SEE coefficients were obtained at primary electron energies of 1 keV. The decreases in the SEE coefficients with temperature were also found for other alumina ceramics, as shown in the Figures 4a and 4b. This tendency agrees with other studies [10-11]. The decrease in the SEE coefficients at high temperature may be caused by the shorter mean free path of the secondary electrons due to the greater phonon and electron scattering in the bulk, thus affecting the shorter escape depth [10]. Therefore, under the high temperature condition, the secondary electrons approaching the surface are fewer than those at room temperature.

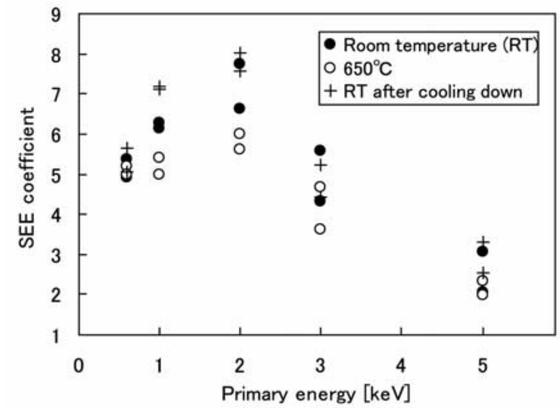

Figure 3. SEE coefficients of alumina ceramics with 92% purity (HA92) as a function of the incident energy at room temperature, 650°C, and room temperature after cooling down.

Figures 4a and 4b also show the relationship between the SEE coefficients of various alumina ceramics and the purity, and the average grain size at room temperature and at 650°C, respectively. According to Figure 4 a, it may be seen that the trend of the SEE coefficients increased with the alumina purity both at room temperature and at 650°C, except for KP990. The SEE coefficients also increased with the average grain size, as shown in Figure 4 b, except for UHA99. Although there are a few exceptions, it is considered that SEE increases with a decrease of the trapping and scattering sites, which may be located in sintering additive materials of the alumina ceramics. The existence of the sites depends on the purity and the microstructure of the alumina.

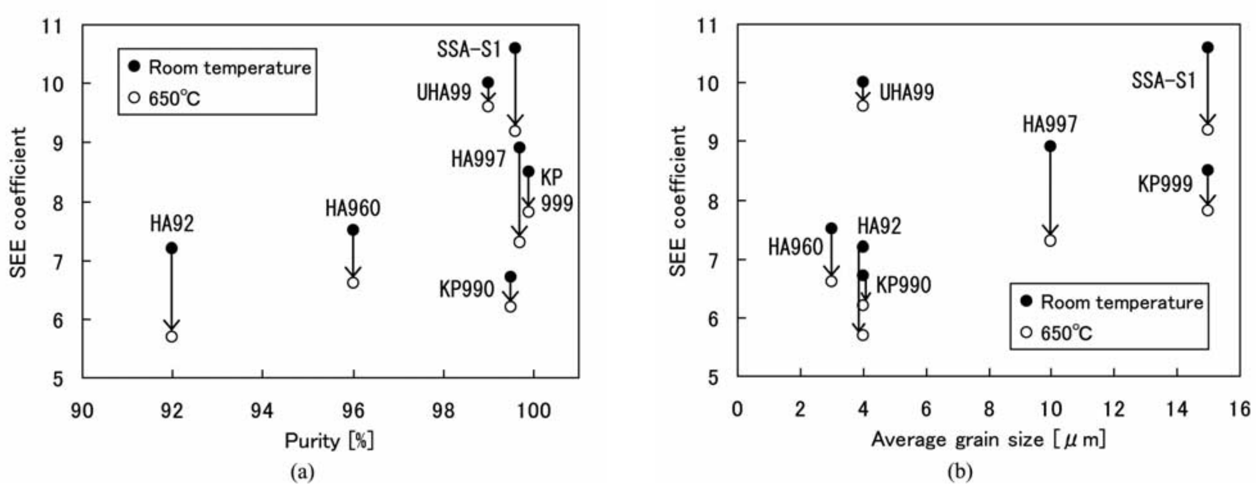

Figure 4. SEE coefficients of several kinds of commercial alumina ceramics at room temperature and at 650°C with the incident energies of 1 keV: (a) relationship between the SEE coefficients and the purity, (b) relationship between the SEE coefficients and the average grain size.

3.1.2 SAPPHIRE

The SEE coefficients of sapphire disk were measured at room temperature and at 650°C. The measurement results are shown in Figure 5. It was found that the SEE coefficients of sapphire increased with the temperature. Since the SEE coefficients after cooling down were almost the same as the previous values at room temperature, the increase in the SEE coefficients at 650°C was caused by the characteristics of the sapphire. This tendency is different from the results obtained for alumina ceramics. Michizono et al. [17] reported that pre-existing F-center defects were found in sapphire, but not in the alumina ceramics examined in this study. Electrons of the F-center which were removed by primary electron irradiations at high temperature probably enhanced the secondary electron creations because the F-center has a state near to the conduction band, leading to easy electron emission [19]. Although the escape depth of the secondary electrons decreased at high temperature, due to an increase of phonon scattering, it is considered that the increase in the secondary electron creation rate at high temperature is dominated in the SEE of sapphire.

3.1.3 SiO_2 AND MgO

SiO_2 and MgO are often used as sintering additives for commercial alumina ceramics. Therefore, it is necessary to measure the SEE coefficients of the materials. Single crystal SiO_2 and MgO disks were examined in this study. The SEE coefficients of the SiO_2 and MgO measured at room and high temperature are shown in Figures 6a and 6b, respectively. According to this figure, it is confirmed that the SEE coefficients decreased with the temperature. This tendency is the same as those of alumina ceramics described before. The decrease was caused by phonon scattering.

3.2 SURFACE CHARGING EVALUATION

3.2.1 MULTI-PULSE MEASUREMENTS

Figure 7 shows the waveforms of the SEE currents of alumina HA960 obtained at room temperature by multi-pulse measurements with patterns A and B. The peak of the waveform decreased with the successive pulses, except for the 4th pulse of pattern B. It is believed that the decrease was caused by positive charging due to the emissions of a large number of secondary electrons from the alumina surface.

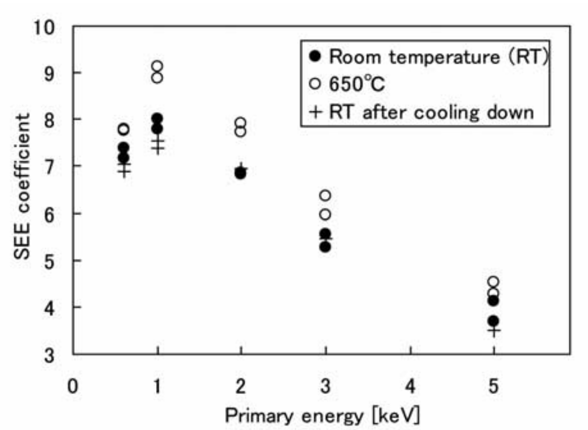

Figure 5. SEE coefficients of sapphire (single crystal alumina) as a function of the incident energy at room temperature, 650°C, and room temperature after cooling down.

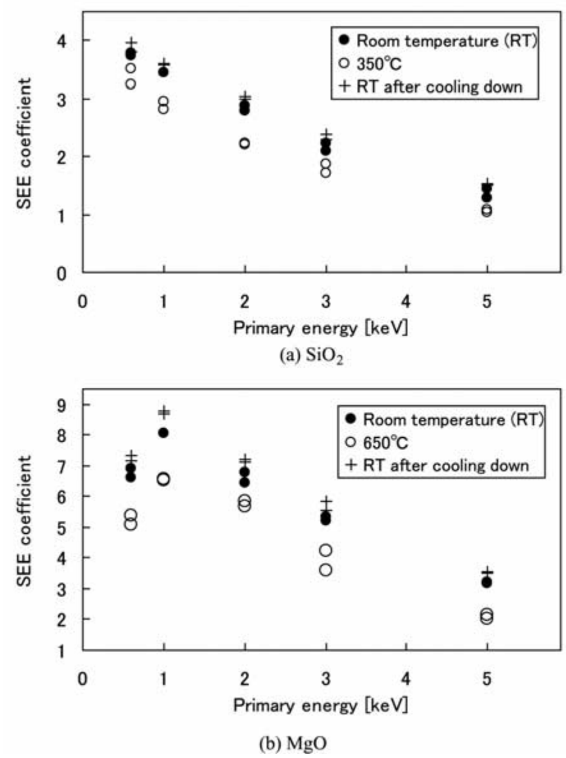

Figure 6. SEE coefficients of single crystal SiO_2 and MgO as a function of the incident energy at room temperature, high temperature, and room temperature after cooling down.

For pattern A, $\rho_{5/1}$ is defined as the ratio of the SEE of the 5th pulse to the 1st pulse, while $\rho_{4/1}$ is the ratio of the 4th pulse to the 1st pulse of pattern B. Here, the SEE was obtained by integrating the secondary currents in each pulse. A typical value of $\rho_{5/1}$ at room temperature was 0.35, which means that the SEE decreased up to 65% due to positive surface charging when 5 successive pulses with 1 ms interval were irradiated. Since the typical value of $\rho_{4/1}$ was nearly unity, surface charging may be neutralized after 4 ms.

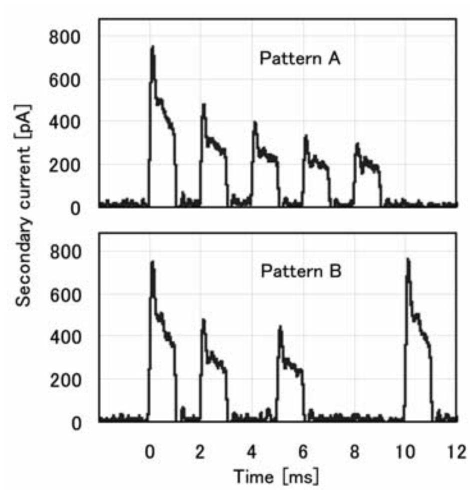

Figure 7. Waveforms of secondary electron currents of HA960 obtained by multi-pulse measurements with "pattern A" and "pattern B" at room temperature.

Figure 8 shows the temperature dependence of $\rho_{5/1}$ of alumina HA960 for the pattern A. It was found that the higher the temperature became, the higher was the value of $\rho_{5/1}$, indicating a lower surface charging. This is probably caused by the higher mobility of the electrons due to high temperature stress.

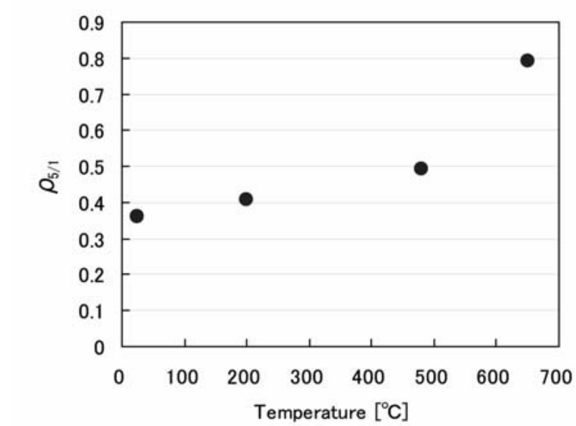

Figure 8. Temperature dependence of the SEE ratio of the 5th pulse to the 1st pulse ($\rho_{5/1}$) of HA960.

3.2.2 SURFACE CHARGING AT ROOM AND HIGH TEMPERATURES

The values of $\rho_{5/1}$ and $\rho_{4/1}$ for several kinds of alumina ceramics, sapphire, and MgO samples at room temperature are shown in Figure 9a. The horizontal axis is the purity. The values of $\rho_{5/1}$ are in the range of 0.23 to 0.58, which means that the SEE became 23% to 58% due to positive surface charging. The higher ratio means that the material has lower surface charging due to higher electron mobility. The surface charging of alumina ceramics and MgO were recovered up to more than 60% after 4 ms, as shown by the values of $\rho_{4/1}$. Sapphire had the lowest values of $\rho_{5/1}$ and $\rho_{4/1}$, and therefore, the largest surface charging compared with others.

Figure 9b shows the values of $\rho_{5/1}$ of pattern A and $\rho_{4/1}$ of pattern B at 650°C. At 650°C, both the $\rho_{5/1}$ and $\rho_{4/1}$ values are around unity, except for sapphire. It could be explained again by the charging relaxation due to higher electron mobility at high temperature.

Obtained values of $\rho_{5/1}$ and $\rho_{4/1}$ for sapphire were smaller than unity even at the high temperature, which means that the electron mobility in sapphire is lower than any other samples. Although sapphire does not have grain boundaries nor additives, a large number of internal defects in sapphire, such as F-centers [17], may play important roles for higher surface charging and poorer charging relaxation.

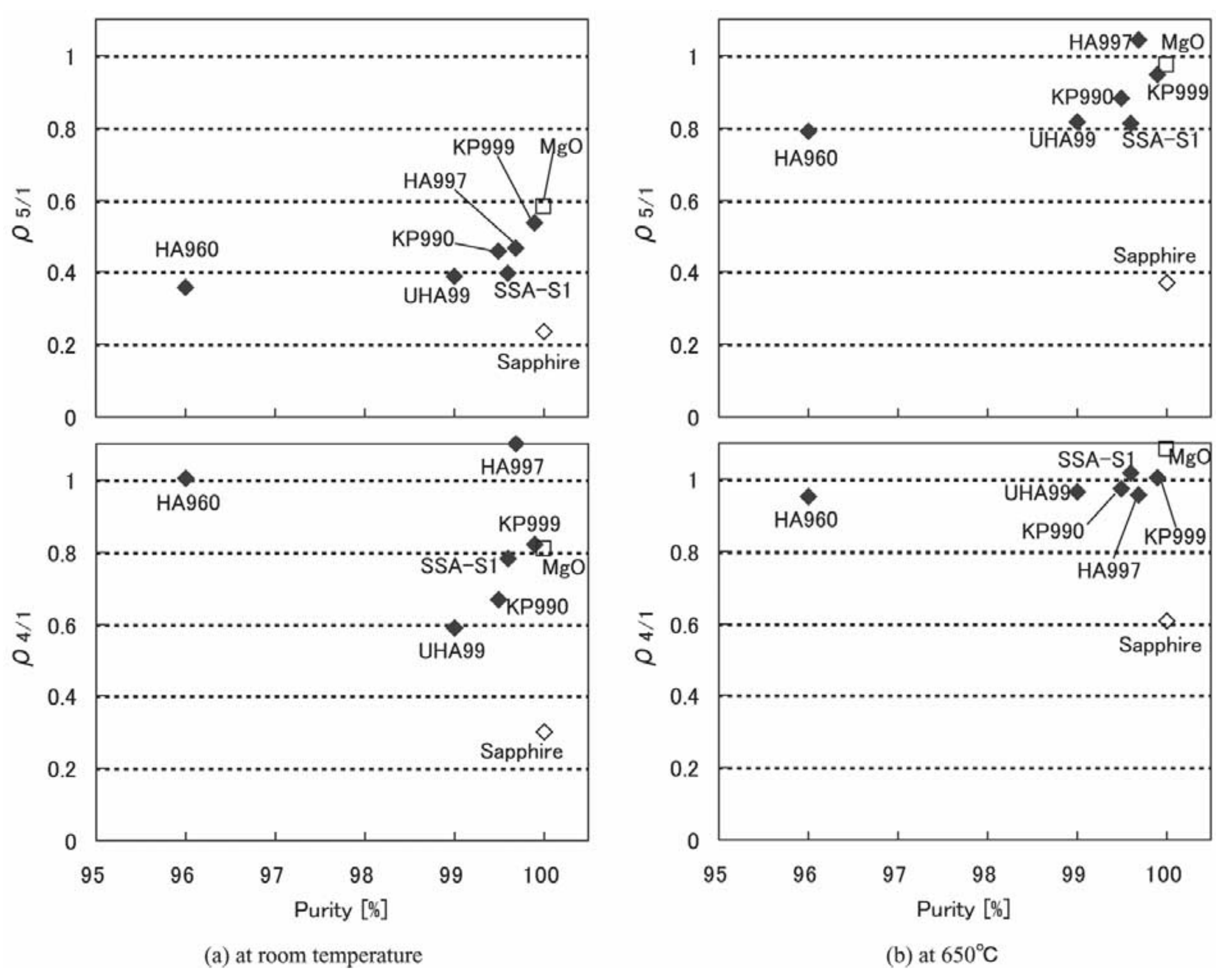

Figure 9. SEE ratio of the 5th pulse and the 1st pulse ($\rho_{5/1}$) and that of the 4th pulse and the 1st pulse ($\rho_{4/1}$): (a) at room temperature, (b) at 650°C.

According to the experiment results obtained in this study it is slightly difficult to understand the relationship between SEE coefficient and surface charging. This is because the surface charging may be affected not only by SEE coefficient, but also other factors, so that the relationship between SEE coefficient and surface charging is not simple.

4 SUMMARY AND CONCLUSION

Secondary electron multiplications (multipactor) and surface discharge are crucial factors that cause the breakdown of an rf window. Since multipator induces localized surface heating, SEE coefficients of commercial alumina ceramics were measured at room and high temperatures. Single-crystal alumina (sapphire), SiO_2 and MgO were also investigated. Decreasesing in the SEE coefficients with temperature were confirmed, except for sapphire. This decrease is caused by the larger phonon and electron scattering at high temperature. The increase in the SEE coefficients of sapphire may be caused by the higher secondary electron creations due to existence of F-center defects.

Surface discharge may result from charge accumulations, which are discharged at the threshold field. Surface charging of the samples was evaluated by multi-pulse beam irradiations. As a result, the surface charging of the samples did not appear at high temperature, except for sapphire. This is caused by the higher electron mobility at high temperature. Sapphire showed higher surface charging than alumina ceramics at high temperature. Pre-existing F-center defects and the higher surface charging in sapphire are considered to be crucial factors that cause its inferior performance compared with alumina ceramics. Further investigation on sapphire is necessary to clarify this consideration. In addition, it was seen that the SEE coefficients of alumina ceramics increased with the purity, except for KP990. The SEE coefficients also increased with the average grain size, except for UHA99. Therefore, the breakdown of alumina rf windows may not be only caused by the SEE coefficients, but also by other properties of the alumina, such as the microstructures, as well as existence of the F-center defects.

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