Industrial Applications of Space Charge Measurement in Japan

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Internal space charge phenomena of insulating materials have been widely discussed recently, and there are many excellent review papers about space charge measuring methods: thermal pulse, thermal step, pressure wave propagation, laser-induced pressure pulse, and pulsed electroacoustic as well as other methods [1-4]. A new task force has been formed with the aim of standardizing space charge measurement in the International Conference on Large High Voltage Electric Systems (CIGRE), Study Committee 15, Task Force 03) [5, 6]. A Japanese working group, the Investigation Committee on Standardization of Space Charge Measurement in Dielectrics and Insulating Materials, also has been formed in the Institute of Electrical Engineers of Japan to examine members’ measurement systems [7]. Although space charge measuring methods are still under development and their performance is still insufficient for many materials, such as semiconductors, we believe they are sufficiently developed to merit a review of this topic. Researchers of dielectric materials and insulation technologies can be considered the main users. While several review papers and a book introduce space charges observed in many kinds of materials, such as polyimide, polypropylene, and fluoropolymers [3, 8, 9], most of the published papers are about high voltage insulation.

In Japan, the pulsed electroacoustic (PEA) method is commonly used for measuring space charge in dielectric materials. Fig. 1 shows a schematic diagram and a test electrode of the PEA method. When a pulsed electric field is applied to a specimen containing space charges, the sudden movement of the charges generates acoustic waves that propagate in the specimen. A piezoelectric sensor under the electrode converts the acoustic waves into electric signals that can be observed by an oscilloscope. The amplitude of the signal is related to the charge density, and the delay indicates the distance from the electrode. In this way, the internal space charge distribution can be observed.

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While there has been a longstanding interest in space charges in cable insulation, recently interest has developed in new materials used in applications such as optical devices, anti-static products, and medical materials.

A basic electrode set-up is shown in Fig. 2. An upper electrode unit consists of the high voltage (HV) electrode and impedance matching circuit that prevents the reflection of high frequency signals. An HV pulse for the measurement and dc bias voltage can be applied by using a coupling capacitor. A piezoelectric sensor under the lower electrode and an amplifier are enclosed in a shielding box. Another amplifier is used in this commercially available set-up. Fig. 3 shows a simple example in which 5 kV dc was applied to a 0.2 mm-thick polystyrene (PS) sheet that had no internal space charge. In this measurement, only surface charges can be observed on both electrodes. The electric field and potential distribution can be calculated easily from the observed charge distribution using Poisson’s equation.

Although the PEA space charge measurement can be applied to many kinds of polymers, about 80% of the published papers discuss polyethylene and polyethylene-based-materials [e.g., 10-35]. One of the reasons for this may be that this research has been supported by electric power companies and cable manufacturers. These companies have measured or have asked uni-
versities to measure space charge for developing and evaluating dc power cables. A dc cable is a good example of an industrial application of this technique; however, the market for dc high voltage cables is disappointingly weak in Japan. Currently, topics on space charge in polyethylene are gradually becoming aimed at fundamental science and are less relevant to real product development.

While there has been a longstanding interest in space charges in cable insulation, recently an interest has developed in other new materials that are used in, for example, optical devices, antistatic products, and medical materials. New clients from different technological fields have visited our laboratory and have repeatedly tried to apply the PEA method to their specimens. Some of the specimens are now commercially available products. Thus, the space charge research field is expanding, as shown in Fig. 4. In this paper, industrial applications of space charge measurement in Japan are described.

In order to demonstrate the new techniques that are becoming available, it is necessary to take into account the rapid progress in the PEA space charge measuring method. Since insulation is likely to be the major interest of readers, the PEA's application to epoxy resin, a common insulating material used in a variety of products from electronic devices to high voltage apparatus, is described as the first example. The next two subjects are concerned with electrostatic phenomena. One is the space charge related to electrostatic discharges (ESD), including initial charges accumulated during a film manufacturing process, and the other is related to charge transport materials designed to move internal charges for photocopiers and new light-emitting devices. Finally, other materials that have already been measured or will soon be measured are presented in the last section.

The PEA method is described here to provide useful information for those who develop many kinds of space charge measurement systems and to introduce new users to this research field.

**Progress of the PEA Method in Japan**

Since the PEA method was developed in 1985 [36] this method has been commonly used in Japan and since then, many researchers have continued to make both fundamental advances and develop optional enhancements, as shown in Fig. 5. A critical performance criterion by which the PEA may be judged is resolution; both the magnitude and spatial resolution of space charge measurement are important. Also of importance is the rate at which the measurements can be repeated. Since the resolution is determined by the signal-to-noise ratio of the whole measurement system, it is essential to reduce the noise both from the outside (e.g., noise induced from the pulse generator) and also from the inside.
(e.g., thermal noise of the amplifier). In addition to these improvements in resolution, several kinds of optional functions have been developed for particular purposes, for instance, high voltage application, temperature control, lighting unit, and useful software for the measurement.

**Improvement of the PEA Method**

**Resolution in the Thickness Direction of a Specimen**

The spatial resolution in the thickness direction of the specimen, i.e., the resolution parallel to the propagation of the acoustic signal, is improved by reducing the width of the acoustic signal. As the acoustic wave is generated by applying a pulsed electric field, the length in time of applied electric pulse must be as short as several ns. Additionally, the acoustic wave is detected by a piezoelectric sensor, so a thin piezoelectric material is required. The spatial resolution was about 100 µm at first and improved to 5 µm in 1994 [37], and the latest value is 2 µm [38]. This was achieved by using an evaporated piezoelectric sensor.

**Resolution in the Surface Direction of a Specimen**

In three-dimensional (3D) measurement of space charge, the resolution in the surface direction of the specimen is also determined. The first 3D measurement was carried out by moving a small electrode mechanically so that the resolution depended on the area of the electrode. This was about 1 mm² in 1996 [39], so that the surface resolution at that time was estimated to be 1 mm. The acoustic lens method for a new 3D PEA system was introduced in 1998; there was also a similar method using the pressure wave propagation method [40]. The lateral resolution of the latest 3D PEA system is about 100 µm; the depth resolution is 5 µm.

**Time Resolution**

PEA measurements may be repeated with high repetition rates for signal averaging purposes and observing transient phenomena. When using a conventional mercury switch it is possible to observe the signal every 400 ms [7]. The interval of the output signal is improved to 20 µs by using a fast semiconductor switch for a pulse generator [41-44]. Thus, it has become possible to measure the evolution of rapid transients in space charge. The space charge under ac electric fields was first measured by a phase-resolving system that measured the profiles at 20 different phase angles in a cycle of the applied voltage [11, 23, 33, 45, 46]. The space charge profile under ac electric fields at such power frequencies can now be measured directly [47].

**Signal-to-Noise Ratio**

The minimum charge density that the method can detect is about 0.1 C/m², a value that depends on the noise reduction level [7]. Although this only corresponds, typically, to one electric charge per 10¹³ atoms, such a charge would distort the electric field by approximately 5 kV/mm over a thickness of 1 mm.

**Various Arrangements of the PEA Method**

**High-Voltage System**

Space charge measurement of a high voltage cable during a dc breakdown test up to 550 kV was achieved in 1994 by using a new method, as shown in Fig. 6, which applies a pulse to the outer semiconductor in applying a pulse voltage [21, 30]. The specimen was an XLPE cable that had 3 mm-thick insulation. During this test, charge clusters moving from one electrode to the other were observed and named “packet
charge." The charge behavior was recognized in many other cases by using sheet specimens under high electric fields, and the phenomena have been considered to relate to the breakdown. Additionally, a large electrode set-up shown in Fig. 7 was developed for a sheet specimen, and it was tested under a high voltage of about 150 kV [17, 29, 30].

**High-Temperature Systems**

The dielectric characteristics of polymers (e.g., mobility and trap sites) depend on temperature, so that it is worthwhile to measure the space charge characteristics at various temperatures. For a sheet specimen, a test electrode in a silicone oil bath shown in Fig. 8 has been used to heat the specimen [18, 31, 48, 49]. A current transformer has also been used to measure the cables under the same condition as that used for common ageing tests of power cables [20]. The basic electrode set-up is small enough to place into a common environmental chamber, where experiments may be carried out at 70°C and 85% RH [50]. Tape or band heaters are a simple (if crude) way to examine the specimen.

**Lighting Unit**

Among the practical functional polymers, photoconductive materials are widely used in copiers and new electro-luminescent displays. Manufacturers of these products have wanted to measure the internal space charge while the specimen is illuminated. In order to achieve this an organic photoconductor has been measured using a modified electrode that has a window on the top with a light bulb as a light source. Recently, an optical fiber has also been employed to supply visible rays of the desired wavelength.

**User Interface**

The output signals of the PEA method must be deconvoluted to obtain the space charge profile. This numerical analysis was initially thought to be complicated. With the progress of software technology, the analysis has been greatly simplified. For several years, data analysis software has been used that could perform the analysis after the measurement [51]. By using more advanced signal processing software, it is now possible to observe the space charge, electric field, and potential profiles in real time. The user interface of the space charge measurement system has also been progressively improved. For instance, people who have never used an oscilloscope have measured a complex time-dependent space charge evolution of a photoconductive polymer. A basic set-up shown in Fig. 2 is now commercially available.

**PEA in Conjunction with Other Measurements**

The improvements mentioned above are only for the space charge measurement system. Since the PEA method is nondestructive, it is possible to subject the specimen to other analytical methods to obtain more information. With respect to charge transport, it is desirable to measure conduc-
tion current during the space charge measurement [41, 52]. Thermally stimulated current (TSC) has been used for a long time in this research field, so that a comparison between the results of TSC and space charge has been done [53-55]. In addition to the electric properties, chemical and physical analyses such as X-ray microanalysis are useful to find the origin of the internal space charge [10, 17, 27, 33, 45, 56].

**Examples of Industrial Applications of Space Charge Measurement**

**Epoxy Resins for Electronic and High-Voltage Insulation**

While an electronic circuit may work under low voltage additions, strong electric fields may exist in thin insulation, as shown in Fig. 9, and good insulation performance is required due to downsizing demands [56, 57]. The cost restrictions of the products might be more than in the case of high voltage apparatus, and chemicals for washing materials are restricted due to ecological concerns. Thus, the quality of raw materials is deteriorating. For example, epoxy resin for a printed wiring board (PWB) often includes many kinds of ion impurities, which can be detected by simple ion chromatography [56]. They are used at fields < 1 kV/mm dc at high humidity and at high temperature. When the insulation is thinner than 20 μm, as in the case of semiconductor devices, it is very difficult to observe the space charge profile under

![Fig. 9 Electric field strength applied to various apparatus and devices](image)

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![Fig. 10 Space charge and copper distributions within a printed wiring board during an ageing test. (White spots in the bottom figures indicate the presence of copper observed by EPMA.)](image)

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![Fig. 11 Initially accumulated charge in a PVC film](image)

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an electric field because of the limited resolution of the PEA method. In the case of power electronic devices, however, there are many kinds of products with insulation about 150 μm thick in which space charge can be easily observed. In addition, the research on insulation is not a primary subject in the development of electronic devices, in contrast to high voltage apparatus. It can be said that scientists in the space charge research field have an opportunity to enter this significant market.

It is well known that ion migration occurs before the breakdown of a printed wiring board, but the word "migration" usually implies "surface migration," which shortens the circuit on the board [59]. In a metal-based printed wiring board, migration occurs inside the insulation. Fig. 10 shows an example of copper ion migration in a PWB. X-Ray microanalysis (EPMA) indicates the presence of copper. Before the voltage application, migration did not appear and the positive charge was observed at the anode. After ageing at 1.25 kV, 85°C, 70% RH, copper ions migrated into the resin, then positive charges also shifted toward the cathode, depending on the ageing time. The space charge measurements indicate the highly conductive area corresponds to the area of the copper [60, 61]. When an excellent 3D space charge measurement system is developed, surface and internal migration will, ideally, be monitored simultaneously.

Epoxy resin is also an important insulation for high voltage apparatus, though to date, few researchers have examined the internal space charge. Since epoxy resin is commonly used with a high content of fillers, it was thought that the acoustic signal might be distorted. Space charge profiles of a common type of epoxy resin were, however, clearly measured. The recent results have revealed that filler and water are the main factors in moving the space charge in a particular type of epoxy resin [50, 62, 63]. The same phenomenon was also determined by a conventional dielectric test method [64, 65]. One of the biggest manufacturers of gas-insulated switchgear (GIS) has just begun to measure the space charge behavior in a thick epoxy resin, and the results of this work will be published.

**Space Charges Related to Electrostatic Discharge**

Electrostatic phenomena are closely related to space charge. Some plastic sheets, polyvinyl chloride (PVC) in particular, show quite high measured values of surface potential, and electrostatic discharge (ESD) easily occurs on those kinds of materials.

If the charge accumulates only on the surface, as in the case of contact charging, it can be eliminated by using an ionizer that supplies ions to enhance recombination of the accumulated charges. If the charge accumulates inside the
polymer, it cannot be easily reduced. Fig. 11 (1) shows the change of charge profiles of an as-received PVC sheet. Negative internal charge was observed initially only on one side, about 100 µm from the surface, and positive charges were induced at the both electrode surfaces. When the surface of the specimen was removed with sandpaper, the space charge distribution at various depths could be clearly observed, as shown in Fig. 11 (2 - 6) [66]. This result suggests that once the initial space charge accumulates inside the polymer during manufacturing process it is stable; the surface potential would thus be maintained and an ESD could occur easily. Printing companies were interested in this initially accumulated charge and suggested that the initial charge accumulation depended on the sheet manufacturing process. Space charge measurement may then be useful to examine the manufacturing processes of various plastic products. For example, an ionizer manufacturer uses space charge measurement to assess the performance of its products [67, 68].

It is well known that ESD causes problems in electronic circuits, resulting in the breakdown or malfunction of many products. In particular, digital communications technology is severely affected by the ESD signal [69, 70]. To prevent charge accumulation, many conductive polymers have been developed. Almost all of them include conductive fillers such as carbon black, metal fibers, and metal powders. Conductive polymers are not able to prevent ESD completely, however. If a charged particle approaches a conductive polymer, it may attract charges to the surface and ESD may occur.

Some kinds of new antistatic polymers have been developed. Among them are polymer solid electrolytes, such as polyethylene glycol (PEG) [71-74]. They have a resistivity in the order of $10^{10}$Ω·cm, which is higher than conventional polymers that contain conductive fillers. According to the manufacturers, additive ions can move in the polymer when hydrophilic polymer solid electrolytes are used. It was expected that internal space charge behavior could be observed easily, and the results provided useful information about their anti-static performance. Fig. 12 shows space charge distributions of a new anti-static polymer (500 µm thick) under an application of 2 kV [66]. The base resin of this material was polymethylmethacrylate (PMMA), and the polymer was alloyed with hydrophilic gum micro-particles that included potassium ions. When the content of the gum micro-particles was 10%, no significant internal charge profile appeared. A signal due to internal space charge was detected at 20%, and positive internal charge accumulated near the cathode at 30%. This is the critical point at which the gum micro-particles begin to form a percolation network inside PMMA. Thus a positive charge is considered to move along the gum micro-network [75, 76]. On the other hand, negative charges were not observed in this specimen. According to the manufacturer, the negative ion is much larger than the positive one; thus the negative charge could neither move nor be localized sufficiently to be detected. The electric field of accumulated positive charge should compensate for the applied electric field [77]. Thus, this new anti-static polymer can keep the surface potential at almost zero if some charges exist on the surface, and it can also prevent the occurrence of ESD.

Fig. 13 shows a space charge profile of another type of anti-static polymer whose base resin was polystyrene.
Charges of both polarities could move under an electric field [78]. These new anti-static polymers are mainly used in electronic device factories to prevent ESD on their manufacturing lines.

**Charge Transport Materials**

As described earlier, copier manufacturers use space charge measurement to investigate organic photoconductive materials used in their products that are designed to use internal charges. An organic photoconductor (OPC) is made of several layers: a charge generation layer, a charge transport layer, an ion conductive layer, and an electron conductive layer. The charge generation layer provides charges when irradiated with a light or rays, and the charges move through the charge transport layer [79-83].

Charge behavior before and after irradiation was clearly observed in a modified specimen that consisted of a charge generation layer (CGL), a charge transport layer (CTL), and a hole trap layer (HTL) [84]. These three layers were placed between two polyester (PET) films, as shown in Fig. 14. Fig. 15 shows space charge profiles that were changed by voltage application and irradiation. When a dc voltage was applied to the specimen, peaks due to charges on both electrodes were observed. A small peak was also detected at the interface near the charge generation layer. After the specimen was illuminated, a positive charge appeared at the HTL, and the positive charge remained after being short-circuited and after applying a dc voltage of the opposite polarity. This experiment indicated that the positive charge was trapped at the hole trap layer by irradiating the specimen.

A more fundamental experiment investigated the charge injection from the anode to the charge (in this case, hole) transport layer [85]. Fig. 16 (a) shows a specimen consisting of a CTL painted on a PET film and various kinds of vacuum-evaporated electrodes. Since the PET film's conductivity is generally low, charges transported through a CTL are expected to accumulate at the interface between the PET film and the CTL. When a dc voltage (5 kV) was applied to the specimen, positive charges accumulated at the interface and remained after the electrodes were short-circuited. As shown in Fig. 16 (b), the accumulated positive space charge and induced negative charges on both electrodes were clearly observed. Each peak is due to interface charges and has a width that gives the spatial resolution of the measurement system, about 10 mm in this case. Since the actual interface charge density \( q \) can be calculated by integrating the volume charge density \( \sigma \) over the width of the peak, Fig 16 (b) presents space charge characteristics using graphs whose ordinates have two scales. The top of each peak of space charge distribution shows its interface charge on the second axis. The interface charge densities obtained by integrating the interface charge distribution curves are plotted in Fig. 16 (c) against the Fermi levels of the electrodes measured using low-energy electron emission spectroscopy [86]. The apparent Fermi level of the CTL was
also measured and shown. According to these experiments, the amount of injected charge depends on the difference between the Fermi levels of the anode and the CTL; the largest charge injection occurs when the Fermi levels are the same.

Based on a study of organic photoconductors, space charge measurement has been applied to organic light-emitting diodes to investigate suitable electrode materials, although only a few papers have been published on this subject. This new device is expected to be used in a new electroluminescent display system [87, 88].

![Graph showing current vs. temperature](image1)

**Fig. 17** Current characteristics of a polyethersulfone film observed in the same manner as that of TSC measurement without poling voltage application

![Graph showing space charge profiles](image2)

**Fig. 18** The space charge profiles of an as-received and heated polyethersulfone specimens

**Other Examples of Industrial Applications**

Several engineering plastics have been investigated for use as new insulating materials in products such as cables in a naval ship and in a nuclear plant. Polyethersulfone, thermoplastic polyimide, polyetheretherketone, and polymethylpentene have been carefully investigated by measuring TSC, conduction current, and space charge [53-55, 89]. Fig. 17 shows current characteristics of a polyethersulfone film, measured in the same manner as TSC measurement without poling voltage application. The spontaneous current as shown in a peak of the 1st run curve was only observed in the as-received specimen. Once the specimen was heated as shown in the 2nd run, the spontaneous current disappeared as shown in the 2nd run curve. Depending on the result, it has been considered that the spontaneous current was caused by an initially accumulated charge during the manufacturing process of the specimen. Fig. 18 shows the space charge profiles of as-received and heated specimens as being the same as those shown in the 1st and 2nd run in Fig. 17. The profiles were measured without applying dc bias voltage, so that they show the charges stored in the specimens. Internal positive space charge only appeared in the as-received speci-

![Graph showing space charge behavior](image3)

**Fig. 19** Space charge behavior of an epoxy adhesive
men shown in Fig. 18 (a). This result confirmed that the spontaneous current was due to an initially accumulated charge in the specimen. Similar results were obtained in thermoplastic polyimide. Manufacturers may find these results useful in the research, development, and improvement of their products.

Since polypropylene (PP) is an important material for a capacitor, it has been investigated in the same manner as polyethylene [90-92].

The following materials have yet to be studied in detail, but have shown interesting phenomena.

Polyvinyl chloride is one of the most common polymers, but its electrical properties under a dc electric field have not been extensively studied. One report on the space charge and conduction current suggests that the presence of a stabilizer prevented internal charge from being accumulated in PVC [93].

It is well known that even glass for electrical use contains a high concentration of ionic impurities used, for example, for reducing the melting point. When dc voltages were applied to several kinds of glasses, huge peaks due to internal charges were detected, except for water-free silica glass [94]. Protonic conduction in glasses has been discussed by physicists for about 10 years [95-97], and space charge measurement would contribute to this research field.

It has been observed that epoxy adhesive generates charges while hardening, as shown in Fig. 19 [98]. Positive charge was generated during the hardening, but the change of space charge became inactive after it solidified. This result confirms that, at least, the gel can be measured by the PEA method. Since gel-like materials used in medical and biological products contain ions that make them functional, they are expected to show interesting internal space charge behavior.

Investigation of space charge behavior of surface-treated polymers has begun by using plasma-treated PET sheets [99] and a corona-charged ionomer. Since ionomers include ions, they will be interesting specimens for space charge measurement [100, 101].

Many researchers for different companies and organizations are now active in space charge research in Japan. In the near future, a highly advanced measuring system, such as a rapid 3D system, will be developed, so that more materials may be observed. Along with the progress of the development, users' desires also may spur further research. Thus, if users have any measurement difficulties, their problems would provide good suggestions for the people who develop the measurement system.

Conclusions

Industrial applications of space charge measurement using the PEA method were introduced. The materials are not only insulating materials affected by internal charges but also new functional polymers that effectively use internal charges. Users of the space charge measurement have had the method advanced to obtain suitable experimental conditions for a variety of materials. The space charge research field has been continuously expanding in Japan, and there are still many prospective materials for space charge measurement specimens.

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References


