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Electrets

Editor: G. M. Sessler

G. M. Sessler Introduction

G. M. Sessler Physical Principles of Electrets

J. van Turnhout Thermally Stimulated Discharge of Electrets

B. Gross Radiation-Induced Charge Storage and Polarization Effects

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S. Mascarenhas Bioelectrets: Electrets in Biomaterials and Biopolymers

G. M. Sessler and J. E. West Applications

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Electrets

Edited by G. M. Sessler

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With 205 Figures

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Preface

Electrets have, over the past decade, emerged as invaluable components in an ever increasing number of applications. Their usefulness is responsible for the recent impressive growth of research work in a field which had been actively investigated since about 1920.

This volume aims to present the fundamental aspects of electret research as well as a detailed review of recent work in this area. The book is broad in scope, extending from the physical principles of the field to isothermal and thermally stimulated processes, radiation effects, piezoelectric and pyroelectric phenomena, bioelectret behavior, and, last, but not least, to applications of electrets. The emphasis of the experimental work discussed is on polymer electrets, but work performed on other organic substances, notably biomaterials, and on inorganic materials, such as ionic crystals or metal oxides, is also reviewed.

The interest in polymer electrets is due to the fact that these show extremely good charge-storage capabilities and are available as flexible thin films. In the 1960s attention focussed on highly insulating polymers, such as polytetrafluoroethylene, which have deep traps that store charges for extremely long periods of time. Around 1970, discovery of the strong piezoelectric properties of polyvinylidenefluoride attracted the imagination of many researchers and an enormous amount of work was devoted to the investigation of the physical and chemical properties of this and similar materials. Today, very active research is underway on charge-storage properties of both classes of polymers.

The chapters of this book are generally self-contained in the sense that each can be understood on its own. There are, however, many cross-references between chapters which will help to guide the reader to related or supplemental material in other parts of the volume. Uniform symbols and abbreviations are employed for the most-frequently used quantities and polymer names. A list of polymer names will be found in Chapter 1, a partial list of symbols at the end of the volume.

Although there have been a few monographs on specific topics of electret research and a number of conference proceedings, a cohesive treatment of the entire field of electrets has so far been lacking. The present volume, by covering many aspects of the field in a relatively small space, is an attempt in this direction. We realize, however, that a number of important questions are not, or not sufficiently, discussed, and that the views held by the different contributors are not always congruent.

It is with great pleasure that the editor expresses his gratitude to his fellow contributors, each being a renowned authority in his field, for their collaboration. The preparation and updating of the manuscripts placed a considerable burden on these colleagues, which they carried with understanding.

The book is dedicated to Professor *Bernhard Gross*, himself a contributor, by his fellow contributors. *Bernhard Gross* is the nestor of electret research, both theoretical and experimental. Apart from this, he has enhanced the knowledge in many other parts of physics. Without his contributions, electret research would not be what it is today. It is with admiration and gratitude that his coauthors devote this book to him.

Darmstadt, September 1979 *Gerhard M. Sessler*

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1. Introduction

G. M. Sessler

With 2 Figures

An electret is a piece of dielectric material exhibiting a *quasi-permanent electrical charge.* The term "quasi-permanent" means that the time constants characteristic for the decay of the charge are much longer than the time periods over which studies are performed with the electret.

The electret charge may consist of "real" charges, such as surface-charge layers or space charges; it may be a "true" polarization; or it may be a combination of these. This is shown schematically in Fig. 1.1 for a dielectric plate. While the true polarization is usually a frozen-in alignment of dipoles, the real charges comprise layers of trapped positive and negative carriers, often positioned at or near the two surfaces of the dielectric, respectively. The electret charges may also consist of carriers displaced within molecular or domain structures throughout the solid, resembling a true dipole polarization. If the charges are displaced to domain boundaries they are referred to as Maxwell–Wagner polarization. On metallized electrets, a compensation charge may reside on the electrode, unable to cross the energy barrier between metal and dielectric. Mostly, the net charge on an electret is zero or close to zero and its fields are due to charge separation and not caused by a net charge.

An electret not covered by metal electrodes produces an *external electrostatic field* if its polarization and real charges do not compensate each other everywhere in the dielectric. Such an electret is thus in a sense the electrostatic analogue of a permanent magnet, although electret properties may be caused by dipolar *and* monopolar charges while magnetic properties are only due to magnetic dipoles. The existence of an external field and the corresponding analogy with a magnet has often been used to define the electret.

However, as *Heaviside* already realized in 1892 [1.1], the fields of an electret may be compensated within a short time period by the relative motion of real charges and dipoles. This is observed in many piezoelectric substances. If one prefers to include some of these materials in the electret category, as *Heaviside* did, it is necessary to use the broader definition of the electret introduced above based on the *permanency of at least one of its charge components* and waive the necessity of an external field. However, such a broad definition includes the entire class of piezoelectric substances. We shall in the following adhere to this definition but include from the group of piezoelectric materials only the polymeric substances in the discussions in this book (see also Sect. 1.3).

While the classical electrets were made of *thick plates of carnauba wax* or similar substances, present electret research frequently deals with *thin-film*

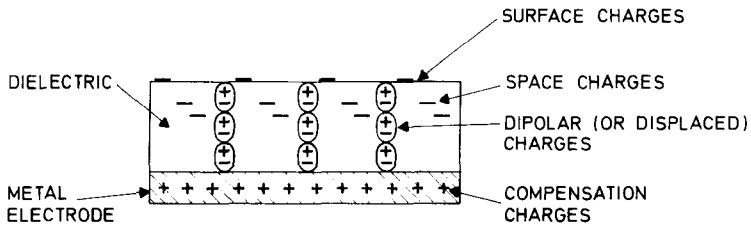

Fig. 1.1. Schematic cross section of a one-sided metallized electret having deposited surface charges, injected space charges, aligned dipolar charges (or microscopically displaced charges), and compensation charges

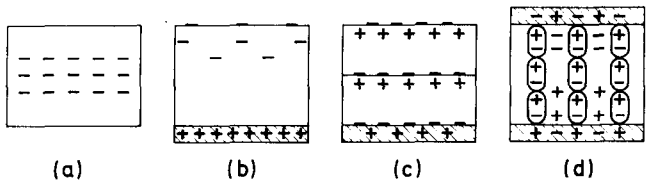

Fig. 1.2a–d. Schematic cross section of some typical electrets without electrodes or with grounded electrodes: **(a)** nonmetallized monocharge electret, **(b)** one-sided metallized electret with surface and space charges, **(c)** one-sided metallized electret with surface charges and charges displaced within domains (Maxwell-Wagner effect) and **(d)** two-sided metallized electret with dipolar and space charges

polymers such as the Teflon materials polyfluoroethylene propylene (FEP) and polytetrafluoroethylene (PTFE) or polyvinylidene fluoride (PVDF). Typical electrets in present use are 10–50 μm thick films of a few cm^2 area, often coated on one or both surfaces with evaporated metal layers. The materials are polarized to charge densities of 10^{-8}–10^{-6} C cm^{-2}, the charges being trapped real charges in Teflon and predominantly dipolar charges (but additionally trapped charges) in PVDF. In the latter case, the dipolar charges are statically (but not dynamically) compensated by space charges due to conduction in the material.

Examples of various nonmetallized and metallized electrets are shown in Fig. 1.2a–d. The charged dielectrics in these figures are either real-charge or dipolar electrets or a combination of these. While the nonmetallized and one-sided metallized samples usually exhibit external and internal electric fields, the fields of the two-sided metallized samples are completely contained within the dielectric.

1.1 Remarks on the History of Electret Research

Electret properties were already described by *Gray* in 1732 [1.2] when he mentioned the "perpetual attractive power" of a number of dielectrics, in particular waxes, rosins, and sulphur. He had generated the static electricity of

these materials by contact electrification caused by cooling of the melts in iron ladles. More than a century later, in 1839, *Faraday* [1.3] theorized about electret properties due to application of an external electric field when he referred to a "dielectric which retains an electric moment after the externally-applied field has been reduced to zero". The word "electret" was then coined in 1892 by *Heaviside* [1.1].

Systematic research into electret properties began in 1919 when the Japanese physicist *Eguchi* formed electrets from essentially the same materials used by *Gray* by a *thermal method* consisting of the application of an electrical field to the cooling melt [1.4, 5]. He found that the dielectrics exhibited charges on their two surfaces which changed sign after a few days from a polarity opposite to one equal to that of the adjacent forming electrode. The charges were later named "heterocharges" and "homocharges", respectively, indicating their relation to the forming electrodes.

In the following decades, electrets from wax materials and a number of other substances were produced by *charging techniques* different from *Eguchi*'s thermal method. One such process, pioneered by *Selenyi* [1.6] in 1928, depends on the *injection of electrons or ions* into insulators. Subsequent extension of this work toward development of electrostatic recordings with powders [1.7] and later investigations of photoconductive image formation by *Carlson* [1.8] culminated in the development of xerography in the 1940s.

Related investigations of the *effect of light* on photoelectric layers by *Nadjakoff* [1.9] in 1938 remained largely unrecognized. *Nadjakoff* studied the charge separation occurring in such layers by simultaneous application of light and an electric field, thus discovering the photoelectret. The effect of photoconductivity on internal polarization was later extensively studied by *Kallmann* and co-workers [1.10, 11] and by *Fridkin* and *Zheludev* [1.12].

In the 1950s, a number of charging methods depending on the application of *high-energy ionizing radiation* were developed. Simplest among these is the bombardment of a dielectric with an electron beam of range smaller than the thickness of the dielectric [1.13, 14].

While *Selenyi*'s work with electron beams was mainly intended for recording, these investigations were performed to study breakdown and thermal charge release. Other similar methods are based on irradiating suitable dielectrics with ionizing radiation, such as penetrating gamma rays or electron beams, and applying an electric field to separate charge carriers generated by the radiation [1.15]. Even without an electric field, charging with gamma irradiation is possible due to Compton currents in the dielectric, as has been shown by *Gross* [1.16].

During the development of xerography, a simple charging method related to *Selenyi*'s ion-beam technique but depending on the application of a *corona discharge* was used [1.8, 17] and later extended to thin films [1.18]. Dielectrics were also charged by application of a *magnetic field* and heat to a dielectric [1.19]. More recently, the charging of thin films with low-energy *electron beams* [1.20] and *liquid contacts* [1.21] was reported.

While the early period produced a significant amount of experimental data, an understanding of charge storage and charge decay was initially not achieved. The first step toward an *explanation* of these phenomena was *Mikola*'s observation [1.22] in 1925 that two charges of different nature and of opposite polarity are found on nonmetallized dielectrics charged by application of an external voltage. One of the charges was assumed to be an internal polarization caused by ionic displacement while the other charge was ascribed to a deposition of ions onto the surfaces. *Adams* [1.23] in 1927, in an attempt to explain the charge decay theoretically, postulated again the presence of two charges, both being due to internal phenomena. These were assumed to be a volume polarization and a "compensating" charge at the surface, the latter being induced by the field of the former. The charge reversal and eventual decay was then explained as being due to the slow decay of the volume polarization and the time lag in the corresponding decay of the compensating charge. Further investigations were made by *Gemant* [1.24] in 1935 who confirmed the polarity reversals found by *Eguchi* and introduced the terms "heterocharge" for a charge of polarity opposite to that of the adjacent forming electrode and "homocharge" for a charge of equal sign.

Convincing evidence for the presence of *two different types of charge* and identification of these charges was eventually offered in a series of fundamental studies by *Gross* [1.25, 26] in the 1940s. He extended the experimental knowledge by measurements of polarization and depolarization currents and charges at varying temperatures and by sectioning experiments. It was demonstrated that the heterocharge is linked to dielectric absorption involving dipoles in polar substances or ionic charges in other materials while the homocharge is due to interfacial charging between electrode and dielectric. *Gross* also derived many of the fundamental relationships governing the fields and charges in a dielectric and the adjacent air gaps and electrodes. The two-charge theory was subsequently used by *Swann* [1.27] as the basis of a phenomenological theory of open-circuit charge decay, extended later on by *Gubkin* [1.28] to short-circuit conditions.

In 1937, *Gross* [1.29] first used the *Boltzmann–Hopkinson superposition principle*, valid for linear dielectrics, to develop a mathematical formalism for the discharge of electrode charges over the internal resistance of the dielectric and for the decay of a polarization in the dielectric. Later, *Wiseman* and *Feaster* [1.30] obtained further extensive confirmation of the validity of this principle by demonstrating that the response of a dielectric to a series of polarization steps is a linear superposition of the individual responses. *Perlman* and *Meunier* [1.31] then applied the principle to the open-circuit decay of unshielded dielectrics and thus explained the charge decay in carnauba wax.

Further insight into the nature of charge retention was achieved from the studies of *Gerson* and *Rohrbaugh* [1.32] which indicated that *carrier trapping* could play an important role in electrets.

Dramatic progress was made by the introduction of the *thermal depolarization method*, first used by *Randall* and *Wilkins* [1.33] for the in-

vestigation of phosphorescence, into electret research. In 1964, this method was applied to the case of dipole polarization by *Bucci* and co-workers [1.34, 35] who suggested the name ionic thermal conductivity (ITC). It allows determination of the activation energy and dipolar relaxation time from a measurement of the depolarization currents obtained upon linear heating of the dielectric. A host of recent work has been devoted to the application of ITC methods or related thermally stimulated current techniques for investigating dipolar and space-charge phenomena respectively [1.36–41]. These studies have culminated in a very comprehensive treatment by *van Turnhout* [1.42].

Of similar importance are recent studies of *isothermal charge transport* in insulators, taking into consideration the effect of trapping on excess-charge currents [1.43–49]. This work has also been extended to irradiated materials [1.50] and materials with non-uniform conductivity [1.46]. Additional information has been gainel from dc and ac conductivity measurements on poorly conducting materials and from the interpretation of such data [1.47, 48, 51, 52]. Recently, charge transport in amorphous solids has been discussed in terms of a stochastic transport model [1.53]. This model describes the dynamics of a group of carriers executing hopping processes in an electric field with a wide distribution of hopping times. Such analyses with and without consideration of trapping phenomena yield a number of important conclusions about charge transport.

Much of the newer experimental work was done on *polymer electrets* which, although chemically not as well defined as many inorganic materials, have gained importance due to their usefulness in practical applications.

Piezoelectric properties of biological and polymeric materials were already investigated by *Fukada* and others in the 1950s and 1960s [1.54]. Of particular importance was the discovery by *Kawai* in 1969 of a strong piezoelectric effect in the polymer PVDF [1.55]. Due to a host of actual and potential applications of piezoelectric polymer materials, this subject has now developed into a very active area of research [1.56–60].

Apart from the use of charge-storage phenomena in xerography and a few other areas, where extreme permanence of the polarization is not required, *applications of electrets* came only recently. Actually, long-lived electrets were generally considered a scientific curiosity until the late 1960s when, a few years after the description of the first polymer-film electret microphone in 1962 [1.61], such transducers were introduced commerically on a large scale. Presently, work on the use of electrets in dosimeters, transducers, pyroelectric detectors, prosthetic and switching devices, gas filters and other instruments is being performed by a number of laboratories [1.62–67] and a series of commercial applications have been reported (see Chap. 7). This has stimulated a considerable increase in very recent electret research.

The early *literature on electrets* was reviewed by several authors (e.g., [1.68, 69]), notably by *Fridkin* and *Zheludev* [1.12] and *Gross* [1.70], while the more recent work is discussed in a number of articles of different scope (e.g.,

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