

Ferroelectrets: Soft Electroactive Foams for Transducers

After certain cellular polymers are internally charged, they behave like soft and sensitive piezoelectrics that can be used to interconvert acoustical or mechanical signals and electrical signals.

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Picture yourself sitting in a fast car, boat, train, or plane and hearing almost no noise. Suppose that you and friends are in your house, apartment, or office and that the living or working space can unobtrusively determine where everyone is. Imagine that the poster or projection screen on your wall can play music and that the pad on your desk can hear you and your visitors. These and similar visions might soon come true with internally charged cellular polymer foams—a novel class of soft electro-mechanical transducer materials that can be used in sensors and actuators.

The word “material” stems from the Latin *materia*, which means “the wooden part of a tree.” An etymological relative is *mater*, that is, mother. Wood is not only one of the most ancient and still most widely used materials, it is also a natural cellular material. Cellular materials (or foams) consist of an interconnected network of solid matter that forms a space-filling structure of open or closed cells.¹ They are naturally abundant, occurring in such substances as cork, sponge, coral, and bone. Artificially made foams are produced on a large scale, yet they are neglected in most undergraduate and graduate physics or chemistry textbooks. They are researched, understood, and documented much less than many other materials. Consequently, opportunities abound for the creative scientist to dream up novel structures and applications.

Nature uses cellular materials to expand the properties of solids, and thus allows for applications that cannot be implemented with uniformly dense solids. Wood, for example, has high stiffness despite its being lightweight. Engineers exploit the foam concept for the same reason. Because they have a wide range of properties, foams can be used for mechanical, thermal, and electrical insulation of almost anything from microelectronic devices to hazardous materials. Foams can be made from polymers, metals, ceramics, glass, and even composites. You often find polymeric foams in daily life. One example is the dispos-

able cups used for hot coffee. The low stiffness of polymeric foams (see box 1) makes them ideal for a wide range of applications in mechanical damping; elastomer foams are the standard material for seat cushions.

Magnet analogues

Permanent magnets such as the ones used to attach a note to a refrigerator are usually based on the ferromagnetic state of certain metals, in particular iron (hence the prefix “ferro”), nickel, or cobalt. In a ferromagnet, the magnetic dipoles order in parallel to give a permanent magnetic polarization, or magnetization, which displays hysteresis in response to a periodic magnetic field of sufficient strength. In analogy, electrically insulating materials whose electric dipoles order like the magnetic dipoles in a ferromagnet are called ferroelectrics, even though they are usually not made from iron. In ferroelectrics, one observes a permanent electric dipole polarization and hysteresis of that polarization in response to a periodic electric field.

Because the ferroelectric state is based on electric dipoles, it comes as quite a surprise that a completely nonpolar material without any molecular dipoles can behave almost like a ferroelectric. As was discovered over the past several years, that unexpected behavior is possible with a variety of foamed plastics. The trick is to charge the voids by subjecting the plastic to a high electric field that generates inside them many tiny microplasma discharges similar to lightning. The resulting material, which carries positive and negative electric charges on opposite internal void surfaces, is called a ferroelectret.² The electret part of the name comes from the surplus electric charges stored in the voids within which individual charge layers are either positive or negative.³ The behavior of ferroelectrets resembles that of ferroelectrics because the overall properties stem from the huge quasi dipoles that comprise the oppositely charged layers separated by the voids.

The preparation, understanding, and application of the new ferroelectret materials is a truly multidisciplinary undertaking that requires technologies and principles developed or discovered by polymer chemists and physicists, polymer-processing engineers, plasma physicists, materials scientists and engineers, acousticians, transducer specialists, and others. Polymer foams were first made in the 1960s,¹ and by the mid-1970s, theoretical and experimental work had demonstrated piezoelectric-like or pyroelectric-like behavior in internally charged heterogeneous polymer systems—for example, a sandwich of soft and hard layers.⁴

The development of cellular ferroelectrets began around 1990 in Finland, when Kari Kirjavainen and coworkers at Tampere University of Technology and later

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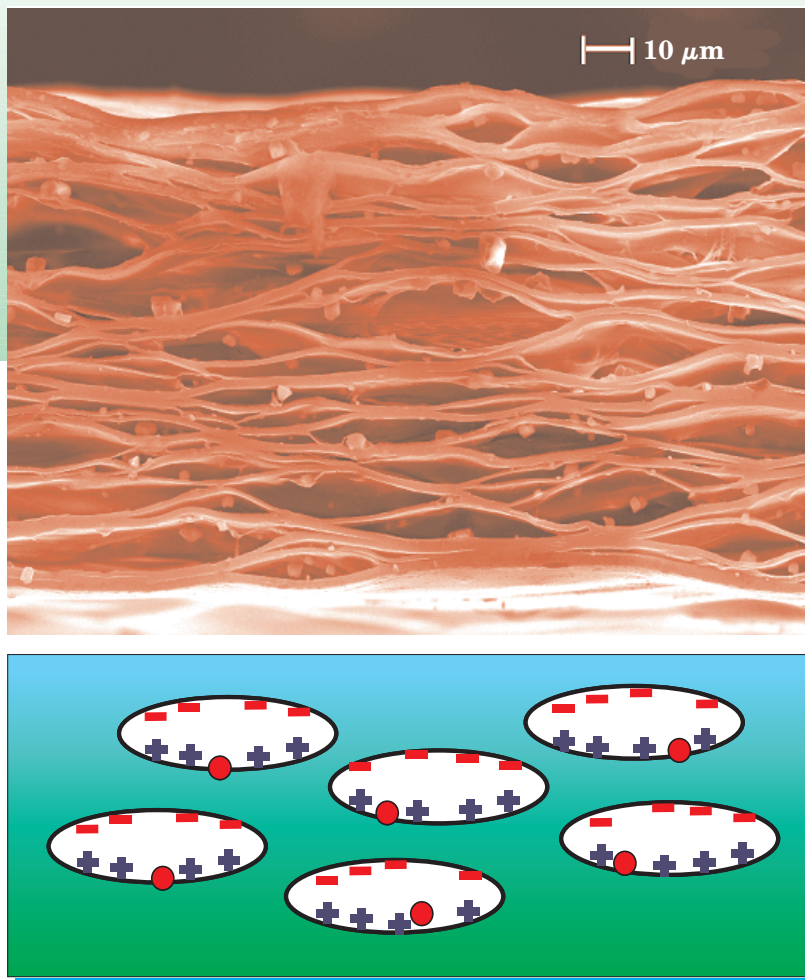


Figure 1. A cellular polymer. The upper image is a scanning electron micrograph of a perpendicular cross section. Grainlike filler particles are visible in the voids. The lower schematic drawing of bipolar voids shows that the tops and bottoms of the voids are oppositely charged. Red circles represent filler particles that facilitate void formation; opposite those particles, the charge density is somewhat reduced.

ize the gas molecules. In addition, flat voids give rather stiff films with reduced electromechanical response. Therefore, to further inflate the voided film, one arranges for the voids' internal gas pressure to be significantly greater than the external pressure. The resulting expansion may be enhanced by softening the polymer through heating.

To obtain electromechanical sensitivity, the voids must be internally charged. Figure 1 shows both a cross section of a cellular film with optimally sized voids and a schematic view of a charged polymer foam. The microplasma discharges that fire as the film is charged can be modified by exchanging the air in the voids with other gases such as pure nitrogen.⁸ An increase in the electrical breakdown strength of the gas in the voids means that the plasma discharge occurs at a higher electric field and that the resulting charge densities are also higher.

How do we know that the rather complex production process really works and that it leads to the desired electric charge layers on the tops and bottoms of the microscopic voids? First, the microplasma discharges in the voids are accompanied by tiny light flashes that can be photographed or even seen with the naked eye.⁹ Second, the charge layers can be inspected with a scanning electron microscope if the voids are cut open at an oblique angle so as to expose their internal surfaces.¹⁰ Because the number of secondary electrons emitted from a surface depends on the surface potential, negatively charged areas appear brighter than the neutral background in an electron micrograph (see figure 2).

Piezoelectricity in cellular materials

For electromechanical or electroacoustic transducer applications, one needs a material with a more or less linear relationship between a mechanical stimulus such as stress or strain and an electrical response, say a voltage or current. Likewise, the material's mechanical response must vary linearly with electrical stimulus. Piezoelectricity can be described by various coefficients that relate mechanical and electric quantities.

In thin ferroelectret films, one of those coefficients is particularly important. Called d_{33} , it specifies the ratio of the charge generated on a thin film to the pressure applied perpendicular to the film's surface. When a voltage is ap-

at the Technical Research Centre of Finland (VTT), also in Tampere, performed the first corona-charging experiments on cellular films made from such standard plastics as polypropylene.⁵ In addition, they reported significant piezoelectric properties of their internally charged films.

Nonetheless, physicists were surprised when in 1999 several research groups presented experimental and theoretical investigations on new internally charged foam ferroelectrets that had large piezoelectric effects. Reports first came out at the International Symposium on Electrets in Delphi, Greece, in September 1999.⁶ Since then, approximately 100 scientific and technical papers about those new ferroelectrets have been published, many of which are mentioned in reference 7. Today a number of commercial and laboratory-scale polymer-foam films are suitable for making electrets, and physicists are coming to appreciate the full range of ferroelectret behavior.

Synthetic cellular polymers

Quite often, voids spontaneously open up in polymers that contain tiny foreign particles such as silicates ("sand") when those polymers are highly stretched. Simultaneous or sequential stretching in two perpendicular directions results in films with lens-shaped voids. Unfortunately, those voids are often too flat for efficient charging by means of internal microplasma discharges because the plasma electrons cannot be accelerated sufficiently to ion-

plied across the film, d_{33} gives the ratio of the change in film thickness to the voltage.

As figure 3 suggests, when a piezoelectric material is compressed or expanded in its thickness direction, its internal dipole moments change in magnitude or density. As a result, compensation charges in the surface electrodes change. Conversely, if a voltage is applied to the electrodes or if extra charges are transferred to them, the dipoles contract or lengthen, or they move closer to or farther from each other. The result is a change in the thickness of the material or in the pressure it exerts on its surroundings.

Classical thermodynamics stipulates a reciprocity of the piezoelectric effect: The coefficient d_{33} does not depend on whether mechanical stress or voltage is applied to the film. A layer model of a ferroelectret predicts that the value of d_{33} is directly proportional to the compressibility of the foam,¹¹ that is, the coefficient is inversely proportional to the foam's Young's modulus. The layer model also predicts the reciprocity of piezoelectric effect demanded by classical thermodynamics.

The charge layers on the internal walls of the foam cells constitute very large electric "dipoles" whose dipole moment can be easily changed by mechanically or electrically stressing the film. The so-called symmetry-breaking length scale associated with those dipoles is quite different from the symmetry-breaking scales of other ferroelectric materials. In conventional crystalline ferroelectrics, a dipole polarization forms spontaneously (see box 2) when the unit-cell dimensions change at the Curie transition. Associated with the transition is a tiny relative displacement of negative and positive ions. In cellular ferroelectrets, the dipole dimensions are given by the thicknesses of the charged voids—usually a few micrometers. In clas-

sical electret transducers or in inhomogeneous layered systems of charged polymer films, charge displacement occurs on a still larger scale given by the thickness of the air gap or of the softer layers.

The macroscopic behavior of the new ferroelectrets is the same as that of traditional piezoelectrics such as quartz crystals or lead zirconate titanate (PZT) ceramics. But the microscopic mechanism leading to that behavior is completely different: In ferroelectrets, the behavior is a result of deformation of charged voids, whereas traditional piezoelectric materials rely on ion displacement in a lattice. Because of the microscopic dissimilarity, the transducer effects of ferroelectrets are often called quasi-piezoelectric, piezoelectric-like, or electromechanical.

The unusual softness and high compressibility of the cellular ferroelectret polymers yields d_{33} coefficients up to 20 times larger than those of the best conventional piezoelectric polymers. Such values are comparable to those of the very hard but brittle piezoelectric ceramics used in many applications.

Ferroelectrets can exhibit thickness resonances analogous to the amplitude resonances of sound in organ pipes. The piezoelectric effect allows an all-electrical, straightforward technique to assess that resonance behavior and to measure mechanical and piezoelectric parameters.¹² Setup and evaluation are taken from dielectric spectroscopy, which is usually used to investigate molecular processes in electrical insulation or in capacitor dielectrics. The spectrum in figure 4 displays frequency-dependent resonances due to electromechanical excitation. As is often the case with such spectra, the figure also shows some higher harmonics. The thickness resonance is broad because of the rather large acoustical damping in the heterogeneous polymer ferro-

Box 1. Elastic Properties of Natural and Artificial Cellular Materials

Elastic properties of various materials are compactly presented by a type of graph called an Ashby materials property chart.¹ The figure at right, which relates Young's modulus to density, is an example of such a chart. Metals, which have high elastic moduli as well as high densities, are found near the top right corner. Fine ceramics are stiffer, but on average a little less dense than metals, while silica glass (not shown) is less dense than most ceramics. Polymers and elastomers have the lowest densities of all solids and are much less stiff than many other materials because of the weak van der Waals bonds between their chain molecules. Polymer-matrix composites, found between polymers and ceramics, combine properties of both classes.

The elastic properties of foams are essential features that enable their use in electromechanical energy conversion. Physicists are still far from a complete understanding of the mechanical properties of foams, so predicting a foam's elastic behavior is a difficult task. For an isotropic open-cell foam, the Young's modulus Y scales according to $Y_0 (\rho/\rho_0)^2$, where Y_0 is the Young's modulus of the solid material and ρ/ρ_0 is the density of the foam relative to the solid. Such simple scaling laws don't work for the strongly anisotropic foams used in electromechanical transducers, which are extremely anisotropic in their elastic properties: Such foams are very soft in the thickness direction and quite stiff in the length and width directions. Typical values for the Young's modulus in the thickness direction are in the range of 1–10 megapascals.

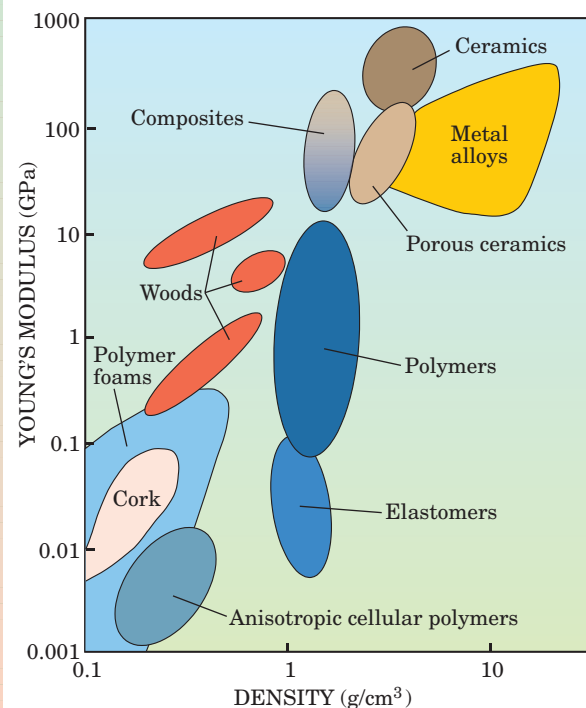
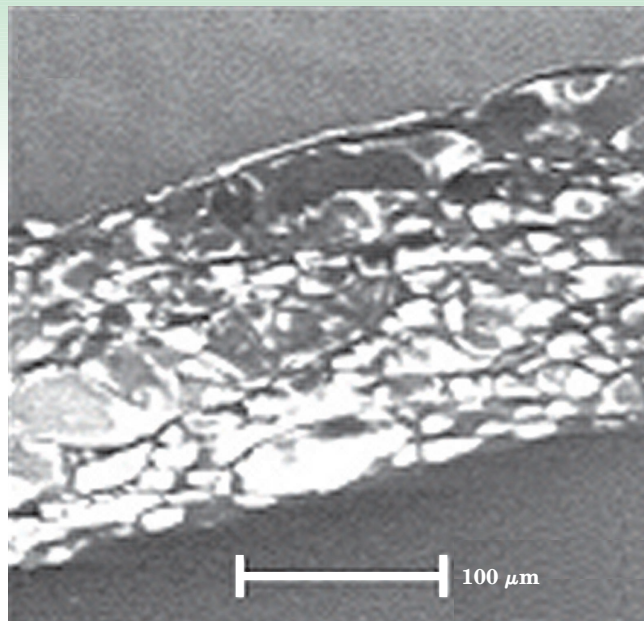


Figure 2. Direct evidence of internal charges in a charged cellular polymer film. The bright areas show secondary electron emission in a cross section that is cut at an angle of a few degrees to the surface. (Adapted from ref. 10.)



electrets—foams are often used as damping materials. From the maximum frequency and the shape of the real and imaginary parts of the dielectric response around the thickness resonance, one can calculate the elastic modulus, the electromagnetic coupling factor k_{33} that relates input and output energy, and the d_{33} coefficient.¹² The frequency response of d_{33} has been investigated over a broad range.¹³ It depends mainly on the Young's modulus and is characterized by high values over a significant frequency range.

A wealth of applications

When compared to traditional piezoelectric materials, cellular polymers have advantages that make them desirable for a variety of transducer applications. Foremost among the favorable properties of ferroelectrets are their large d_{33} coefficients. The table on page 42 lists the piezoelectric coefficient of optimized cellular polypropylene together with the coefficients of some classical piezoelectrics.

Cellular films are thin and lightweight and can easily be made in almost any size or shape. Furthermore, they can be readily handled and are relatively inexpensive. The rather small specific acoustic impedance (the product of a material's density and sound velocity) of voided polymers is an advantage in many transducer applications. The impedance of the polymers is much closer to that of water than is the impedance of ceramic piezoelectric materials. As a consequence, the polymers can emit a larger portion of the acoustic energy generated in an underwater sound source. A significant advantage over most of the classical piezoelectrics is that the majority of piezoelectric cellular polymers consist of nontoxic constituents.

The new ferroelectrets have attracted a lot of interest, but they are not without their problems. Foremost among them is that for some polymer ferroelectrets—including the present workhorse, cellular polypropylene—high temperature causes d_{33} to be permanently reduced. Cellular polypropylene also has limited long-term stability. An ad-

ditional problem with ferroelectrets is that the ideal linear behavior assumed for a good piezoelectric cannot be realized over a very large range of mechanical or electrical loads.¹⁴ That problem exists for all transducer materials, but understanding precisely how it plays out in the new ferroelectret materials will require additional research. Once the underlying phenomena and expected behavior are known, they can be easily accounted for in device applications. The possibility of fatigue must also be considered for ferroelectrets that are used over long periods of time. Scientists in a number of laboratories have begun working to eliminate the problems in ferroelectrets. In particular, efforts are under way to develop a replacement for cellular polypropylene, one that will have similar piezoelectric properties but greater stability.

One property of ferroelectrets may be either a disadvantage or an advantage. Ferroelectrets have a small value for a piezoelectric coefficient called d_{31} , which relates voltage across the sample to the length change in a transverse direction. That small value is a disadvantage in applications that require large deflections due to thermostat-

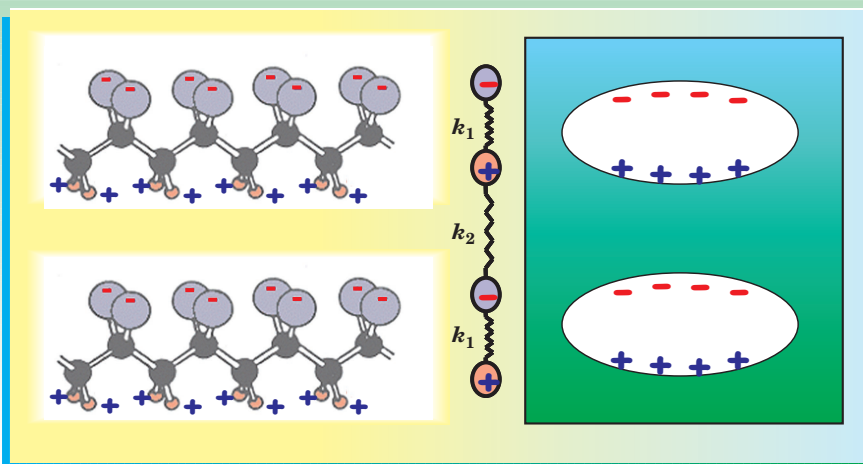


Figure 3. Charge separation in piezoelectrics. The scale of the charge displacement is much smaller in polymeric or ionic ferroelectrics (left) than it is in ferroelectrets (right). A spring schematic (center) illustrates coupling forces inside and between the dipoles. (Adapted from ref. 2.)

Box 2. Hysteresis and Ferroics

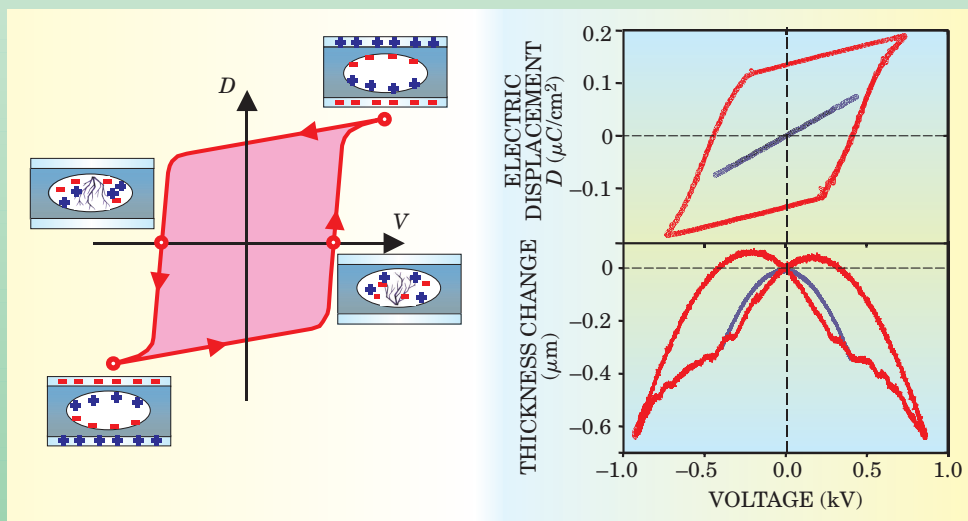
An electric field E applied to matter always induces a polarization P , defined in most textbooks as the dipole moment per unit volume. That definition is suitable for polar polymers, in which microscopic electric dipoles can be identified, but it is much less useful for the description of inorganic crystals.¹⁶ In general, the polarization can be related to an applied electric field by $P = \epsilon_0(\epsilon - 1)E$, where ϵ_0 is the vacuum permittivity and ϵ is the material's dielectric constant. A quantity related to the polarization is the electric displacement, or flux density $D = \epsilon_0 E + P = \epsilon_0 \epsilon E$.

Application of an electric field, however, is not the only way in which a material can be electrically polarized: Polarization can occur spontaneously, without an applied electric field, as in pyroelectric materials. Mechanical stress can also change polarization, as it does in piezoelectrics. Piezoelectric polarization may be less familiar than polarization induced by an external electric field, but it plays an important role in sensors and actuators.

Ferroelectric materials are always pyroelectric (and hence, always piezoelectric), with an important additional feature: A modest electric field—one that will not cause dielectric breakdown—can switch the spontaneous polarization between states (usually two) that are thermodynamically stable and crystallographically well defined.¹⁶ For that reason, ferroelectric materials display hysteresis of the electric polarization as a function of the applied electric field that is described by a so-called hysteresis loop. But hystere-

sis is not limited to polarization: All other physical quantities that couple to the polarization also show hysteretic behavior. The mechanical strain induced by an applied electric field is a particularly important quantity, because ferroelectric materials are always piezoelectric. A wide class of materials, including ferroelectrets, ferroelastics, and ferrofluids, show hysteresis and other behaviors analogous to those of ferromagnets and ferroelectrics. The catchall term for this general class of materials is ferroics.

The figures below show ferroelectret hysteresis loops. The plot on the left indicates the hysteresis of the electric displacement as a function of applied voltage. As the figure shows, during cycling of the voltage the direction of the macroscopic dipoles can switch back and forth because of internal microplasma discharges in the polymer voids. That switching is the essential physics behind the displayed hysteresis. The intravoid microstorms leading to the switching are accompanied by visible, lightninglike flashes.⁹ The right-most figures show two experimental plots, adapted from reference 2. When the cycling voltage does not attain a specific threshold or coercive value, the material does not become charged. As indicated by the blue curves, the electric displacement varies linearly with voltage and the thickness change, which is related to mechanical strain, varies quadratically. The red curves illustrate the hysteresis obtained for voltages that rise above threshold. The thickness variation shows a characteristic "butterfly" loop.



like bending. It is an advantage in applications relying only on the change in thickness of the ferroelectret.

Because of their unique features, piezoelectric cellular polymers can be used in a variety of electromechanical, electroacoustic, and ultrasonic sensors and actuators. Other proposed applications for the new ferroelectrets include thermoelectric converters and underwater transducers. Researchers from VTT and Finland's Emfitech have played a leading role in developing those applications.¹⁵

Usually the device configuration is simple, consisting of a piece of piezoelectric film with an electrode on the top and bottom surfaces. Contacts to the two electrodes complete the design.¹⁴ When the device functions as a sensor, a mechanical or acoustic force applied to one of the sur-

faces causes a compression of the material. Via the direct piezoelectric effect, a voltage is generated between the electrodes. In actuators, a voltage applied between the electrodes causes a strain on the film through the inverse piezoelectric effect. If necessary, one can increase the sensitivity of a sensor or actuator by stacking several layers of the cellular film. Individual layers of the stack can be electrically connected either in series or in parallel. Another modification, appropriate for transducers with distributed sensing or directional characteristics, is to pattern the electrodes, the sign of the polarization, or the polarization magnitude.

Electromechanical transducers usually operate at low frequencies, ranging from zero (direct current) to a few hundred hertz; in that range, the d_{33} coefficients of the new

Piezoelectric Coefficients of Several Useful Materials

Piezoelectric material	d_{33} (pC/N)
Quartz (silicon dioxide)	2 (d_{11})
Lead zirconate titanate (PZT)	170
Polyvinylidene fluoride (PVDF)	20
Cellular polypropylene (optimized)	600

ferroelectrets are very high. Because cellular films can be fabricated in various sizes and shapes, detectors based on those versatile materials can be used in a number of contexts to sense force, impact, vibration, or motion. The range of possible applications includes keyboards, impact sensors, and accelerometers. Cellular-film based detectors are currently used, although on an experimental basis, in biomedical applications for monitoring heartbeat, breathing, and forces on limbs. An intriguing possibility under study is that cellular sensor films might be used in floor mats that track patient movement in hospitals or as components of intrusion detectors around industrial or military installations. Such applications depend on the direct piezoelectric effect. But the inverse effect can also be used in, for example, electromechanical actuators to control motion or displacement.

Foam in the home

Of great interest to engineers are applications of the new piezoelectric materials to microphones, headphones, and loudspeakers. The simplicity and lightness of electroacoustic transducers facilitates their use for many specific tasks in communications, noise control, hearing aids, toys, surround sound, and so forth.

The possibility that cellular polypropylene could be used in speakers was suggested shortly after the discovery of the material's strong piezoelectric activity. Polypropylene-based speakers can be built to operate in a "thickness" or "membrane" mode. For speakers that operate in the thickness mode, a film is simply supported or glued onto a

solid surface and excited with the amplified audio signal voltage. Thickness variations of the film cause a sound wave to be generated and radiated into the surrounding medium. The thickness-mode design, however, generates low sound-pressure levels over the audio frequency range. It is better suited for ultrasonic applications.

The story is different when the speaker operates in membrane mode. In such speakers, the polymer film is situated between two perforated and stiff electrodes, with an air gap on both sides. The polymer film contains charge of a single sign, so that if a signal voltage is applied between the two electrodes, the film is deflected. The sound waves so generated radiate into the surrounding medium through the holes of the perforation. Speakers operating in membrane mode rely on electrostatic forces rather than the piezoelectric effect used for thickness-mode speakers. But they use the same kind of cellular films and produce higher-intensity sound waves—well in excess of 100 dB in the mid- and high-frequency ranges above 500 Hz—than the thickness-mode devices. Other major advantages of membrane-mode speakers are the flatness of their frequency response and their light weight. Membrane-mode transducers could be very appropriate for commercial theater and public-address sound systems and might be applied at home in surround-sound and home-theater systems, or to actively control ventilator noise in ducts and sound transmission through walls.

Cellular-film based piezoelectric microphones are simple devices comprising a ferroelectret film with connectors and housing. They possess a flat frequency response throughout the audio range but at present are somewhat less sensitive than classical electret-type transducers. The cellular-film microphones, however, do not require an air gap. In classical electret microphones, the air gap between membrane and backplate, which needs to be on the order of 20 μm , is a major complication. Thus, if the d_{33} coefficient can be further increased, cellular-film microphones may well conquer a significant part of the microphone market.

Applications of the new ferroelectret films extend well beyond the audio frequency range into the ultrasonic region—up to and including the thickness-resonance frequency usually found between 0.1 and 1 MHz. Ultrasonic

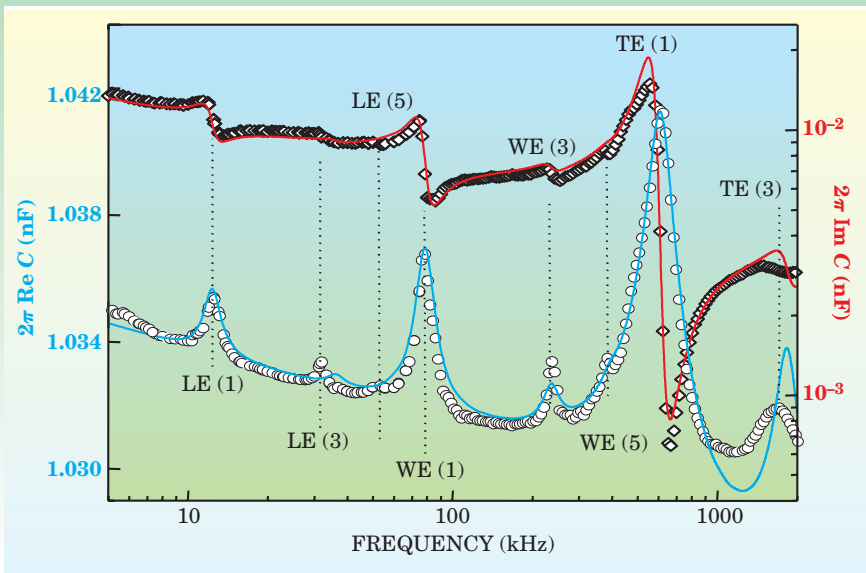


Figure 4. Dielectric spectroscopy as a tool for investigating piezoelectric properties. The spectrum displays both experimental data and overlaid theoretical curves. The vertical axes specify the real (blue curve) and imaginary (red curve) parts of the complex capacitance C . Visible on the spectrum are thickness (TE), length (LE), and width (WE) resonances. In addition to the fundamental (1), the spectrum indicates higher third (3) and fifth (5) harmonics. (Adapted from Neugschwandtner et al., ref. 12.)

transducers based on ferroelectrets can serve, for example, as transmitters of control signals, as transmitters and detectors for burglar alarms, and as transmit–receive devices for distance measurements in robotics or vehicle-distance monitoring.

We expect that in the near future, internally charged cellular polymer films will be integral components of numerous devices that will improve the quality of life by delivering high-quality audio and practically noise-free environments, enhanced security, vital medical information, and more. Picture the possibilities!

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