1'01 .YMER PIEZOELECTRIC TRANSDUCERS FOR UI TRASONIC NDE

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ABSTRACT

Excitation of ultrasonic waves can be done by numerous methods that induce time-dependent elastic deformation or pressure whereas receiving such waves requires means of converting time dependent displacements to an electric signal. Since piezoelectric crystals are offering a low cost, highly effective actuator/sensor materials they remained for many years the leading form of ultrasonic transduction. The most widely used commercial piezoelectric material is the various phases of lead zirconate titanate (PZT). Unfortunately, ceramic transducers are fragile and it is very difficult to produce them in large sizes. Further, these piezoelectric materials have a relatively high acoustic impedance requiring a complex damping and matching techniques to induce broadband signals. Polymer piezoelectric materials have been known to exist since the end of the twenties but the discovery of Polyvinylidene Fluoride, during the sixties, made them useful for commercial ultrasonic transducer applications. Piezoelectric polymers are associated with a low noise and inherent damping that makes them very effective receivers as well as broadband transmitters for high frequencies tasks. This paper reviews pol ymer piezoelectric materials, the origin of their piezoelectric behavior and their applications to ultrasonic NDE.

1 NTRODUCTION -1'1 EZOELECTRIC SENSORS AND ACTUATORS

The piezoelectric effect was discovered by the Curie brothers in 1880. The practical use of piezoelectric materials became possible with Paul 1 angevin's discovery in 1916 of the piezoelectric characteristics of quartz crystals. Following this discovery, it was observed that some crystalline materials demonstrate a spontaneous polarization along one axis of the crystal, ferroelectric behavior. For many years, Rochelle salt was the only crystal that was known to have this ferroelectric propert y.

Progress in this field toward a practical piezoelectric materials for ultrasonic NDE applications became more significant with the discovery of barium titanate, BaTiO3, in 1947 and the ability to activate it as a piezoelectric material by poling. This success was followed by the observation of the very strong effect in lead zirconate titanate (P ZT) ceramics. A brief review of the car] y development of PZT transducers can be found in [1 ,ee, 1990a, and 1990b]. Today, there are several hundreds of known piezoelectric materials. Generally, there is a considerable interest in ferroelectric crystals as transducer materials for their spontaneous polarization and the strong sensitivity that is attributed to their higher electromechanical coupling than piezoelectric crystal ,such as quartz. While ferroelectric materials have a higher electromechanical coupling, they are not as stable as the single crystal piezoelectric materials. Further, both the poly - and the single

crystals are made of brittle materials, which limits their practical size for high frequency ultrasonic applications where thin wafers are required.

For many years, the need for flexible and large area piezoelectric material for ultrasonic transducers was well recognized. The discovery of piezoelectricity in polymers in the early twenties made this possible [Sussner, 1979]. The largest progress in this field was made in the sixties with the discovery of piezoelectricity in Polyvinyl idene Fluoride (PVDF). After this discovery, numerous applications of this polymer were reported and ultrasonic transducers have emerged as practical NDE tasks [Harsanji, 1995]. Since PVDF is still the leading piezoelectric polymer, the emphasis of this paper this material.

PIEZOELECTRIC POLYMERS

The existence of piezoelectric polymers was already known since 1924, however the early known piezoelectric polymers did not receive much attention until the work by Fukada in the fifties and sixties [Fukada, 1964]. Fukada and his co-workers discovered that rolled films of polypept ides and numerous other polymers induce surface charges when st ressed. A major milestone in this field was recorded with the Kawai's discovery of the strong piezoelectric effect in polyvinylidene fluoride (PVDF or PVF2) [Kawai, 1969] in 1969. Later, other PVDF co-polymers were also reported, including P(VDF-TrFE) [Bui, Shaw and Zitelli, 1986] and P(VDF-TeFE) [Tasaka and Miyata, 1985] and others.

Ferroelectric polymers are produced by a variety of techniques, where in the case of PVDF the material is mechanically drawn and polarized in order to form a useful transducer material. The drawing techniques include extrusion and stretching and while processed the film material is subjected to a strong electrical polarization field. Without drawing, PVDF shows a very weak piezoelectric behavior and the higher the molecular orientation tbc stronger the resultant response of tbc polarized film.

After polarization, PVDF exhibits considerably stronger piezoelectric response than most other known polymers [Kawai, 1969]. The discovery of tbc piezoelectric and later of the pyroelectric properties of PVDF and the growing applications of this polymer [Tamura, 1975] sparked extensive research and development activities. Some of the piezoelectric polymers that arc known today include: polyparaxylene, poly-bischloromethyuloxetane (Penton), aromatic polyamides, polysulfone, polyvinyl fluoride, synthetic polypeptide and cyanoethul cellulose [Wang, 1 Herbert and Glass, 1988].

ORIGIN OF 1'1EZOELECTRICITY 1 N PVDF

The origin of piezoelectricity in PVDF is not well understood and the "dipole model" is the most wide] y accepted explanation of its origin. This material is a semi-crystalline high-molecular weight polymer with repeat unit (Cl_2 - CF_2) whose structure is essential] y head-to-tail, i.e., Cl 12

- CF_2 - $(CH_2$-$CF_2)_n$-CH_2 - CF_2. PVDF is approximately half crystalline and half amorphous. The crystalline form consist of a mm-polar α phase and a highly polar β phase (See Figure 1). Where the α phase is a non-polar phase, whereas the β phase has the hydrogen and fluorine atoms arranged to give maximum dipole moment per unit cell. These dipole moments are randomly oriented until the polymer is electrically polarized. Under the application of an electric field, the polymer chains inside the crystallites align themselves along the field by rotating the dipoles around the chain axis. The piezoelectric response of PVDF polymers is the result of a net polarization.

PVDF demonstrates hysteresis loops similar to those known from crystalline ferroelectric materials [Wang, Herbert and Glass, 1988]. As can be seen from "1'able 1, this polymer demonstrates piezoelectric constant, d33, of more than -30 pC/N which is very high. This constant d33 is negative. in other words, applying an electric field in the direction of polarization (film thickness) causes the film thickness to decrease. PVDF has an anisotropic characteristics where in the film plan tbc strongest effect is in the drawing direction as opposed to the normal direction, i.e., d31> d32 > 0. The ratio of these two d-constants can vary between 510 1() times. In Figure 2, the normalized d constant (divided by d33) is plotted as a function of the angle with tbc drawing direction. In '1'able 1, a comparison is given between the properties of PVDF and some of the commonly used piezoelectric materials. While PVDF has a relatively low dielectric constant compared to piezoelectric ceramics and crystalline materials, it has a relatively large value compared to other polymers.

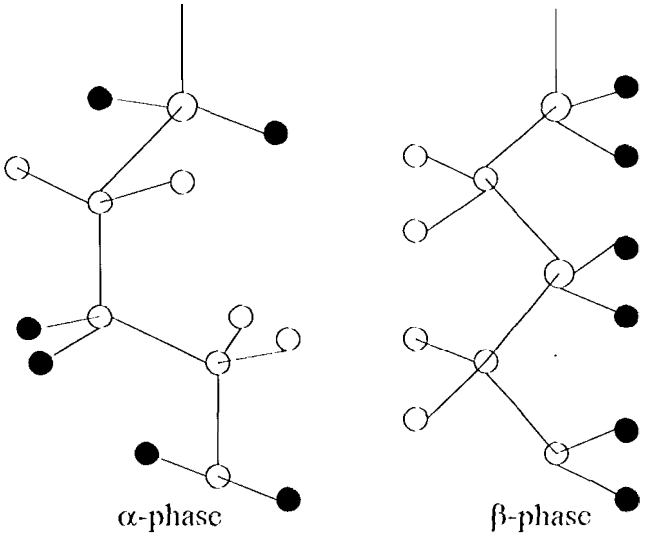

FIGURE 1: Schematic view of two of the PVDF phases. The u-phase consists of a series of non-polar anti -parallel chains, whereas ~-phase phase consists of a series of polar parallel chains.

TABLE 1: Comparison between commonly used crystalline piezoelectric materials and PVDF.

Material	ε	d33 (pC/N)	g33 (1 0-³Vm/N)
BaTiO3	1700	191	'-12.6
Quartz	4.55	2.3(d11)	50.0 (g31)
PVDF	13	-33	-339.0
PZT-4	1300	289	2 s . 1

It is interesting to point out that the piezoelectric coupling coefficient, K33, for PVDF is two times higher than quartz. These two coupling constants, K33 and K31, for PVDF are showing significantly different temperature effects, As can be seen from Figure 3, while the thickness coupling, K33, stays constant around 0.2, K31 drops significantly as the temperature drops below -50 degrees C. The curie temperature of PVDF is near 110 degrees C which makes it useful for some elevated temperature applications. in an applied electric field, the piezoelectric force acts primarily normal to the film direction and its coupling coefficient is independent of temperature. On the other hand, lateral effect is caused by transverse contraction of the film and the value of the Poisson ratio v] 3, is a temperature dependent parameter that varies from 0.2 to 0.6. This Poisson's ratio variation occurs as the material manifests the transition through the glass transition point, 'l'g, from the glassy to the rubbery state. The large Poisson's ratio at the level of 0.6 is related to the anisotropic behavior. The negative value of d33 as opposed to positive d31 and d32 is explained by the Poisson ratio behavior of compression in the thickness direction while expanding in the plane direction.

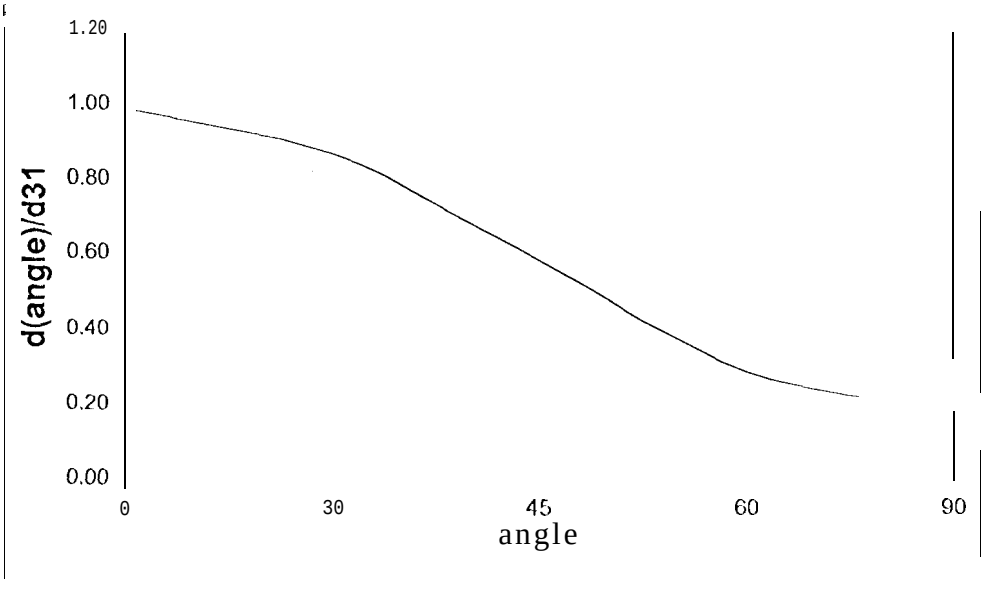

FIGURE 2: Relative transverse d constant as a function of angle with the drawing direction [Fukada and Yasuda, 1 964].

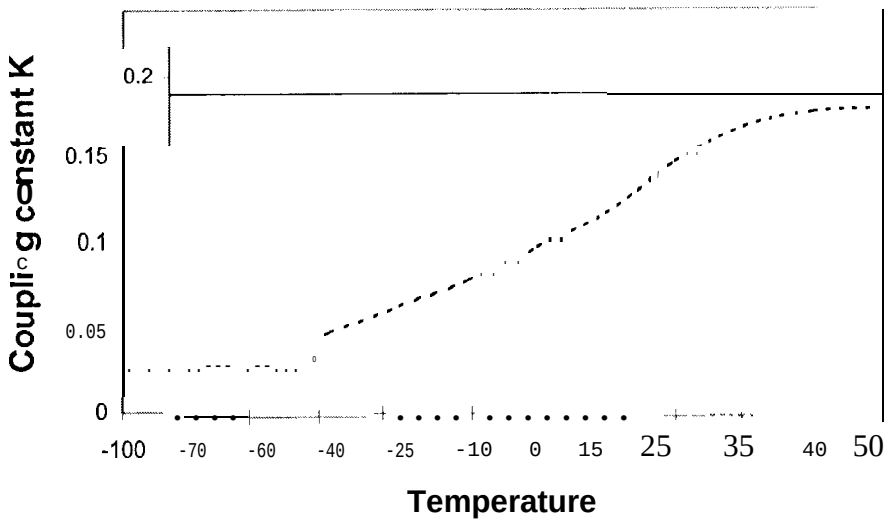

FIGURE 3: Temperature dependence of the piezoelectric coupling constant for PV1)F in the drawing direction (K_{31}) and normal to the film plane (K_{33}) [Ohigashi, 1976].

APPLICATION OF PIEZOELECTRIC POLYMERS FOR ULTRASONIC NDE

In recent years, polymer piezoelectric materials were reported to be applied to numerous fields taking advantage of the flexible characteristics of these polymers. Some of the applications of these polymers include: Audio devices - microphones, high frequency speakers, tone generators, and acoustic modems; Pressure switches - position switches, accelerometers, impact detectors, flow meters and load cells; Actuators - electronic fans and light shutters.

PVDF shows a strong piezoelectric response even at microwave frequencies. The very high g constant of PVDF at the level of-339 x10-3 Vm/N, as compared to 50 x10-3 Vm/N in Quartz, 12.6 x10-3 Vm/N in $BaTiO_3$, and 25 x10-3 Vm/N in PZT, is making this polymer an ideal receiver of ultrasonic signals. PVDF [Fukada, and Yasuda, 1964] is being increasingly used for commercial ultrasonic transducers [Chen and Payne, 1995]. The application of these materials to ultrasonic NDE requires an ability to predict its performance to allow effective design of ultrasonic transducers. Lee and Moon [Lee, and Moon, 1990] have proposed a modal coordinate analysis using laminated PVDF films, whereas Lee investigated the reciprocal relationship between PVDF modal sensors and modal actuators [Lee, 1990].

PVDF was also reported to be used for acoustic emission experiments. Stiffler and Hennecke [1982], used a commercially available transducer and water couplant to monitor the signals related to fatigue tests of composite materials. The transducer responded very effectively to the in-plane displacement components of the acoustic emission in composite materials. The sensor

successfully monitored fatigue loading and the advantage of this application of polymer transducers is the strong fracture toughness of these polymers,

PVDF ultrasonic transducers for broadband NDE applications was reported by several investigators, including [Ohigashi, 1988, Shaw, et al, 1981, Chen, 1978, and Charome, 1 979] Broadband transducers can bc easil y made with such films in the thickness range of 9 to 20 μm and the film low impedance enables a direct coupling to water. The low acoustic impedance of this polymer makes it attractive to medical applications of ultrasonic imaging. The transducer can be coupled directly to the patients skin or eye with minimum discomfort while maintaining an effective sound transmission to the test area, other forms of making PVDF transducers include tbc usc of aluminum backing. Al uminum has a relatively low acoustic impedance as compared to other widely used metals and a direct backing of aluminum enables to form an effective broadband ultrasonic transducer. The flexibility of the polymer allows to fabricate transducers in a wide variety of shapes for special applications.

Conventional ultrasound imaging transducers suffer from the trade-off between bandwidth and sensitivity, which impedes the optimization of ultrasound image quality. Using PVDF transducers allows to alleviate this trade-off by providing an overall high sensitivity while retaining wide bandwidth properties. This is achieved by making usc of the pulse compression technique in a multilayer assembly. A detailed theoretical model that allows the performance of the multilayer transducer to be predicted was developed. Experimental data were collected by using a wideband PVDF needle hydrophone [Zhang, et al, 1993]. A good agreement was obtained between the experimental data and compute] simulations.

PVDF was also reported in acoustic microscopy applications where focusing transducers based on a 9-μm thick PVDF foil were fabricated. The transducer operates in the frequency range of 20- J 60 MHz with 78 M Hz operating frequency in water and provides a lateral resolution of 27.5 μm and a vertical resolution of 35 μm. Using such an acoustic microscope images of transistors and microelectronic components were made using focused PVDF transducer [Smolorz, and Grill, 1995].

PVDF is increasing y being used for array transducers in medical II-scans and some industrial NJ)']' instruments. The construction of tbc multi-element polymer transducer requires a careful attention to the definition of the small array el ements. One of the main benefits of polymer arrays is their low mechanical and dielectric cross talk between the elements. Therefore it is much simpler to prepare such an array as compared to the ceramic ones where dicing is necessary. Photolithography techniques arc used to form the array and to define the electrode pattern. Another advantage of using piezoelectric polymers is the ability to print microelectronic devices, such as preamplifiers, onto the film allowing to produce micro-electro-mechanical devices (MEMS) [Harsanji, 1995].

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