A History of Medical and Biological Imaging with Polyvinylidene Fluoride (PVDF) Transducers

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***Abstract*—Polyvinylidene fluoride (PVDF) is a ferroelectric polymer with unique properties suitable for use in a wide range of medical and biological imaging applications. Most notable among these is its low acoustic impedance, which matches that of the body reasonably well, and its flexible mechanical properties. This paper traces the exploitation of PVDF as a transducer material from its early beginnings for thyroid and breast imaging to its current well-established applications in ultrasound biomicroscopy. Although PVDF's electromechanical properties fall short of composite ceramic materials in the traditional diagnostic frequency range, it has significant advantages in the 25- to 100-MHz range. Design criteria for high frequency transducers are reviewed, and examples of relevant medical and biological images are used to illustrate the excellent image quality obtained with this remarkable material.**

I. Introduction

Since the discovery of the strong piezoelectric properties of PVDF over 30 yr ago by Kawaii [1], researchers have sought to apply this unique material to a wide range of industrial and medical applications. In the medical imaging field, the need to consider alternative materials stemmed from the recognized limitations of conventional ceramics, such as lead zirconate titanate (PZT). PZT has a high acoustical impedance and is difficult to form into focused geometries because of its brittle nature. PVDF, by virtue of its low acoustical impedance and rugged flexible constitution, appeared to address both of these limitations. Indeed, early polymer transducers demonstrating spectacular bandwidths of over 100% were routinely achieved. However, the comparatively low coupling efficiencies of 0.15 to 0.2 of the polymer materials compromised performance in the diagnostic frequency range. The subsequent discovery of enhanced piezoelectric properties in copolymers such as P(VDF-TrFE) [2] improved penetration limits of polymer transducers but not sufficiently to achieve widespread acceptance in the diagnostic frequency range. One of the factors that sealed the fate of polymer ferroelectrics in the diagnostic frequency range was the development of composite ceramic materials [3], [4], which currently dominate the medical transducer market. The composite ceramics currently provide the best compromise among acoustical impedance, bandwidth, dielectric properties, electromechanical efficiency, and signal-to-noise ratio for single-element and array transducers in the frequency range below about 15 MHz. Despite the fact that PVDF has not found applications in imaging at frequencies less than 15 MHz, it is essential for hydrophone construction and remains the standard for ultrasound field calibration systems. The paper by Harris *et al.* in this special issue examines the important contributions of PVDF hydrophones to modern ultrasound imaging [5]. By far, the most successful applications of polymer ferroelectric materials have occurred at frequencies above 15 MHz. Here, PVDF and its copolymers have established a strong presence and have arguably produced the best images of tissue microstructure.

In this paper, the 20-year history of PVDF transducers in medical and biological imaging applications is traced. Discussion of the initial attempts at diagnostic imaging will focus on breast imaging applications. Explorations of the high frequency range (>15 MHz) is discussed in more detail. Most of the example images are taken from our own work, but readers are reminded that the references point to the substantial contributions of many others in this field. Examples of transducer design, fabrication, and evaluation are reviewed. Throughout this paper, images from relevant medical and biological applications are used to highlight the considerable contributions of PVDF and its copolymers to imaging science and medicine.

II. Historical Background

The first literature reports of the use of PVDF in soft tissue imaging were by Ohigashi *et al.* [6] and Foster *et al.* [7] in 1979. Ohigashi's transducer consisted of a 13-mm diameter, 5-MHz single element in which the PVDF was mounted on a spherically concave copper backing. A schematic of the device and an early image of the thyroid is provided in Fig. 1. Good resolution (for the time) was achieved, but penetration was limited to a few centimeters. At the same meeting, Foster *et al.* reported on an unusual geometry in which a cylindrical PVDF transducer

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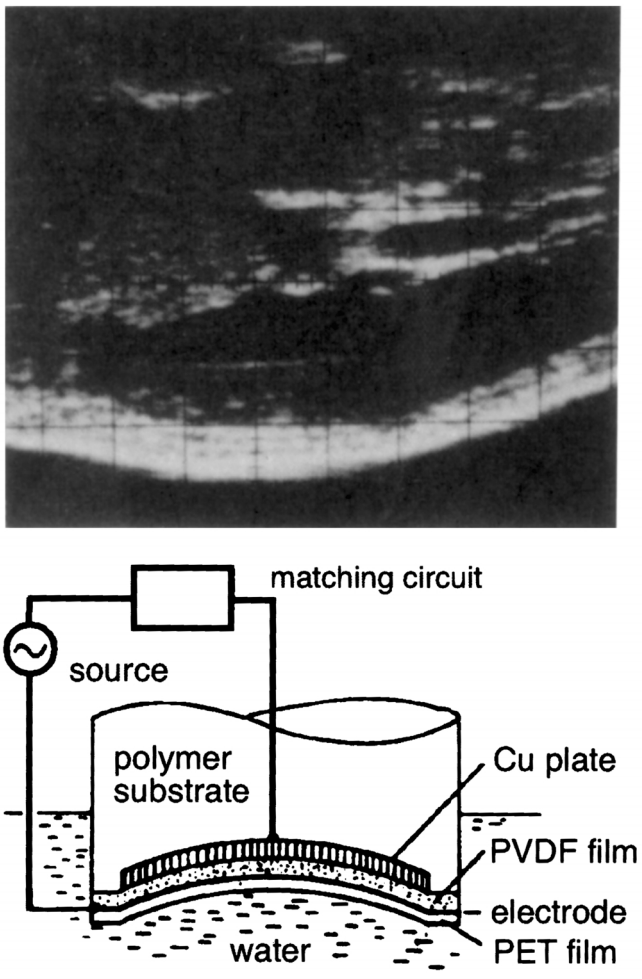

Fig. 1. Schematic of early (1979) PVDF single-element 5-MHz focused transducer (right) courtesy of Ohigashi *et al.* [5] and image of thyroid tissue (left). Bar = 2 cm.

was used as a receiving element while a PZT transducer aimed along the cylindrical axis was used as a transmitter. This hybrid device offered a very large depth of field and high resolution. An image of an ex vivo kidney specimen made at 3.5 MHz using the cylindrical hybrid is shown in Fig. 2. Ambitious attempts were made by Swartz and Plummer [8] to integrate polymer devices into monolithic silicon array structures for ultrasonic imaging. Unfortunately, technological difficulties prevented the successful implementation of these ideas.

The primary application of early polymer transducers was in the area of breast imaging and tissue characterization. In 1984, Foster *et al.* [9] reported the development of a 13-MHz "macroscope" that was used in an extensive study of breast tissue properties. This system relied on a strongly focused 13-MHz polymer transducer that executed quantitative C-scan analysis of excised tissue specimens. Images of ultrasound attenuation and velocity, such as those shown in Fig. 3, were used to characterize breast tissue. The quantitative results in Fig. 4 show an unambiguous segmentation of breast fat, fibroglandular tissue, and infiltrating duct carcinoma. At the time, 13 MHz was

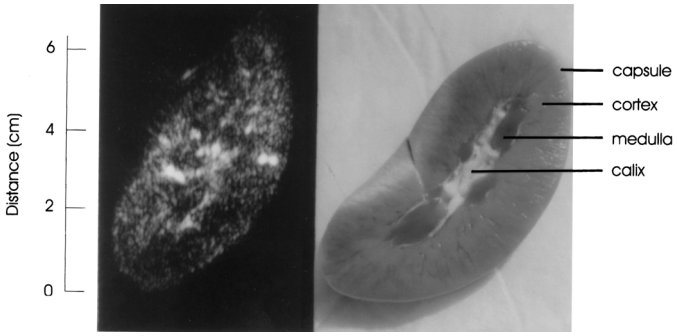

Fig. 2. Cut section of bovine kidney (right) and corresponding image made in 1979 using a novel cylindrical hybrid PZT/PVDF transducer configuration. Image courtesy of Foster *et al.* [6].

considered a very high frequency for breast imaging, but the results of Fig. 4 are actually quite relevant today as breast imaging is now performed using center frequencies in the 7.5- to 15-MHz range.

Adaptations of the hybrid polymer design were exploited in prototype commercial breast imaging systems in the early to mid 1980s. The Life Instruments Corporation (Boulder, CO) breast imaging system, using a combination of conical PVDF and annular PZT transducers, is shown in Fig. 5. Details of the transducer design are also shown in Fig. 5. Note the large conical PVDF element surrounding the inner annular array. This system was used in a clinical study of over 1700 patients [10]. An example image of the breast of a normal subject is given in Fig. 6. Jackson *et al.* [11] collaborated with Labsonics Inc. to develop a 7.5-MHz spherically focused PVDF transducer in their breast imaging system. This transducer provided significant improvement in image quality over lower frequency spherically focused transducers. The Labsonics scanner provided excellent differentiation of fatty and fibroglandular tissue as shown in Fig. 7 (left) and good boundary definition of lesions as shown in Fig. 7 (right). Automated whole breast ultrasound imaging such as that described previously has gradually been replaced by real time hand-held breast examinations performed with PZT linear array transducers. PZT is more suited to array applications by virtue of its high dielectric and coupling constants. Attempts to improve performance of PVDF transducers by means of multilayer and coded multilayer designs have been investigated by Zhang *et al.* [12]. Although these approaches showed promising results, they have not been applied successfully in clinical imaging.

III. High Frequency Polymer Transducers

In medical and biological imaging above 15 MHz, PVDF and its copolymers have proven extremely useful. Whereas ceramic transducers have struggled with sensitivity, reproducibility, beam properties, and fabrication difficulties, polymer transducers have provided simple, compact, rugged, and diffraction-limited performance. An excellent source of reference information on PVDF is the review paper by Brown [13]. Table I provides a comparative list

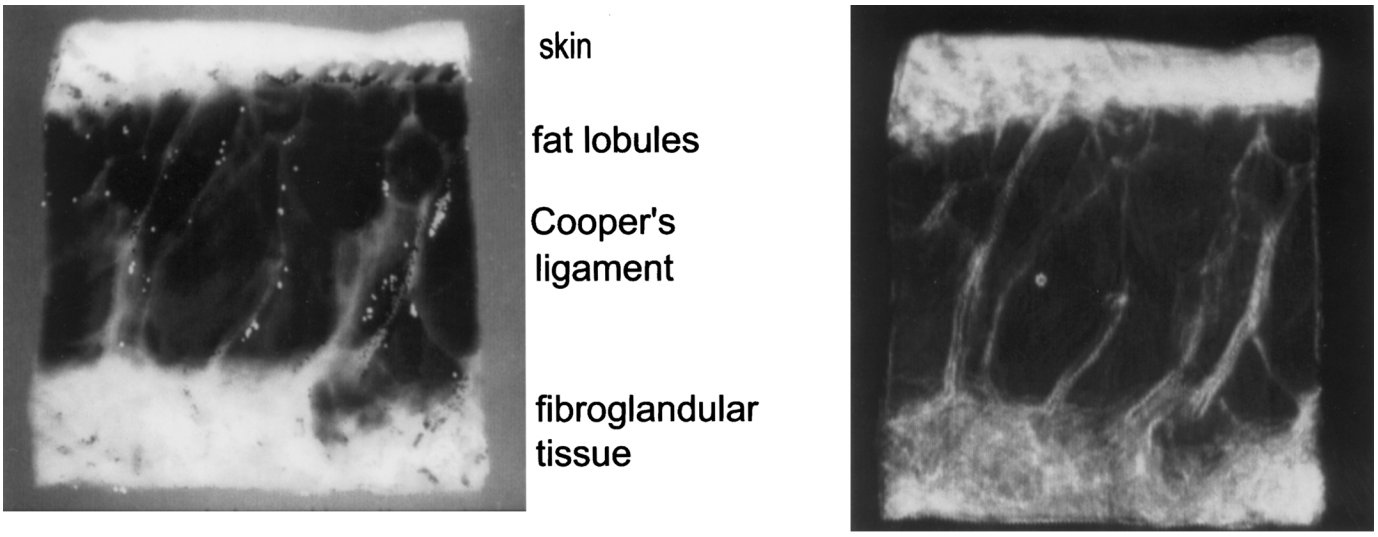

Fig. 3. Quantitative images of breast tissue in vitro using a strongly focused 13-MHz PVDF transducer. Left image: speed of sound (black = 1380 m/s; white = 1636 m/s). Right image: attenuation (0 dB/cm = black; 7.5 dB/cm = white). Images courtesy of Foster *et al.* [8].

TABLE I
COMPARISON OF ELECTROMECHANICAL PROPERTIES FOR PVDF, P(VDF-TRFE), AND PZT5A.

Parameter	PVDF	P(VDF-TrFE)	PZT5A
Sound speed (m/s)	2200	2400	4350
Density (g/cm^3)	1.78	1.88	7.75
Z (Mrayl)	3.9	4.5	33.7
Relative dielectric permittivity	6.0	5.0	1200
Mechanical Q	10	25	75
Electromechanical coupling coefficient	0.15–0.20	0.3	0.49
Mechanical flexibility	Outstanding	Satisfactory	Poor

of relevant material properties for PVDF, P(VDF-TrFE), and PZT5A.

Stark contrast in physical properties between the polymers and the ceramic is clearly evident in Table I. Although several important properties, such as electromechanical efficiency and mechanical Q, are superior for the ceramic, its high acoustical impedance and brittle nature are a major limitation in the high frequency area. At 50 MHz, the required thickness of PZT5A for a resonant structure is 43.5 μm. This means that the active element is only 6 to 8 times thicker than the grain structure of the ceramic. Not only does this cause a decrease in coupling factor and an increase in the electromechanical losses [14], [15], but it also results in a very fragile structure for incorporation into a transducer. Thus, the combination of PVDF's low acoustical impedance, modest piezoelectric properties, and high mechanical flexibility made it an ideal choice for high frequency imaging. Some of PVDF's properties, such as its frequency-dependent dielectric and mechanical losses, required special consideration in accurate modeling of the materials performance. Brown and Carlson [16] reported useful models for the measurement of acoustic and dielectric properties of PVDF in 1989. Fig. 8

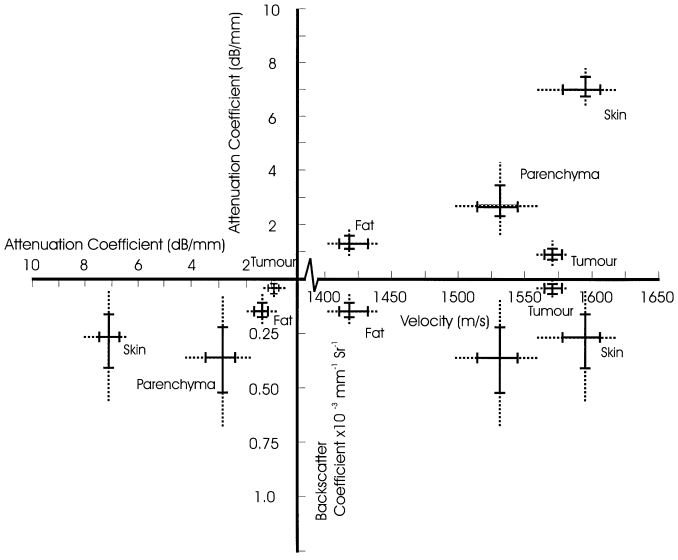

Fig. 4. Measurements with 13-MHz PVDF transducers provided the first detailed examination of breast tissue acoustical properties. In this plot, speed of sound, attenuation, and backscatter are all displayed. Note the unambiguous segmentation of fatty tissue, fibroglandular tissue, and infiltrating duct carcinoma at this frequency. Dotted lines = range; solid lines = standard deviation. Adapted from [8].

Fig. 5. Automated whole breast imaging system manufactured by Life Instruments Corporation in 1983. This system was a prototype employing a hybrid PVDF cone annular array transducer. This scanner was used in a clinical study of over 1700 women at Women's College Hospital, Toronto. The transducer is shown at the right.

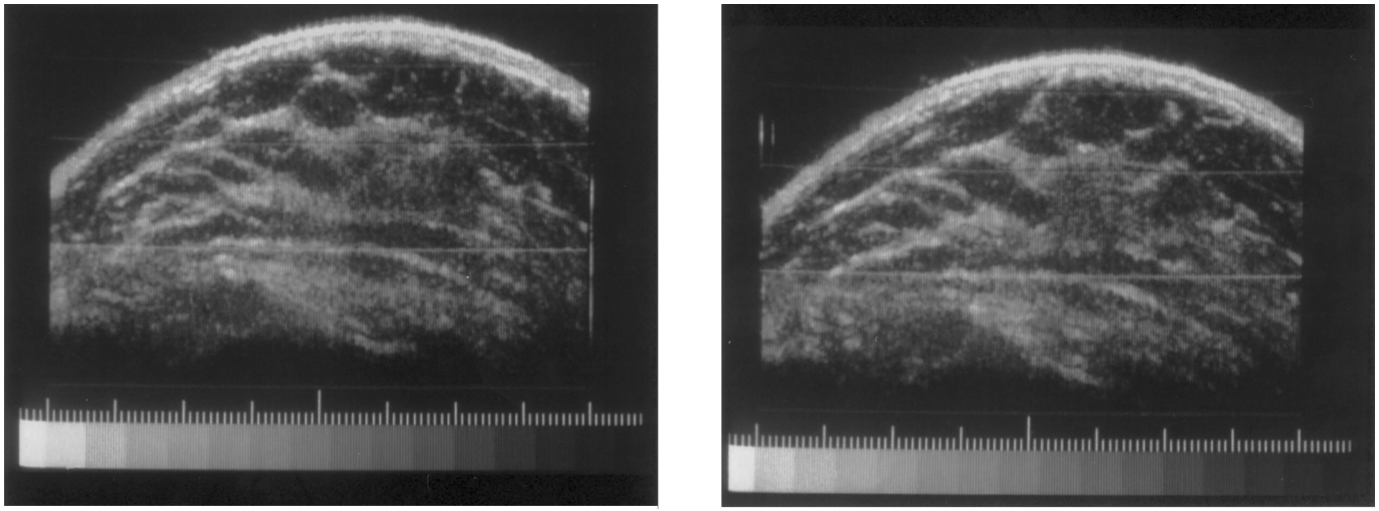

Fig. 6. Example of a breast image made with a PVDF cone/annular array scanner. Note the fine speckle texture and sharp definition of this image at a frequency of 4 MHz. Images courtesy of Foster *et al.* [9].

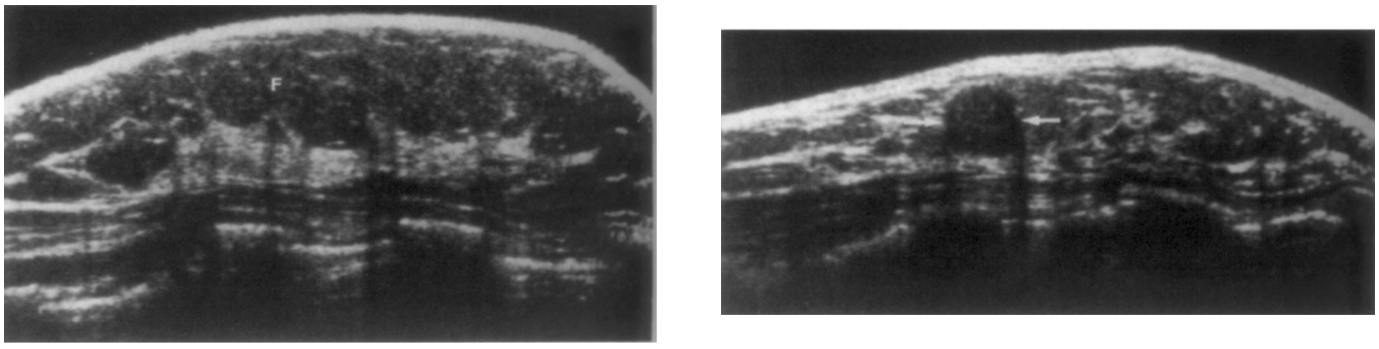

Fig. 7. Outstanding breast images were also possible using polymer transducers in the pulse-echo mode. Here a 7.5-MHz focused P(VDF-TrFE) transducer is used to make an image of normal breast tissue (left) and a breast lesion (arrow, right). Images courtesy of Jackson *et al.* [11].

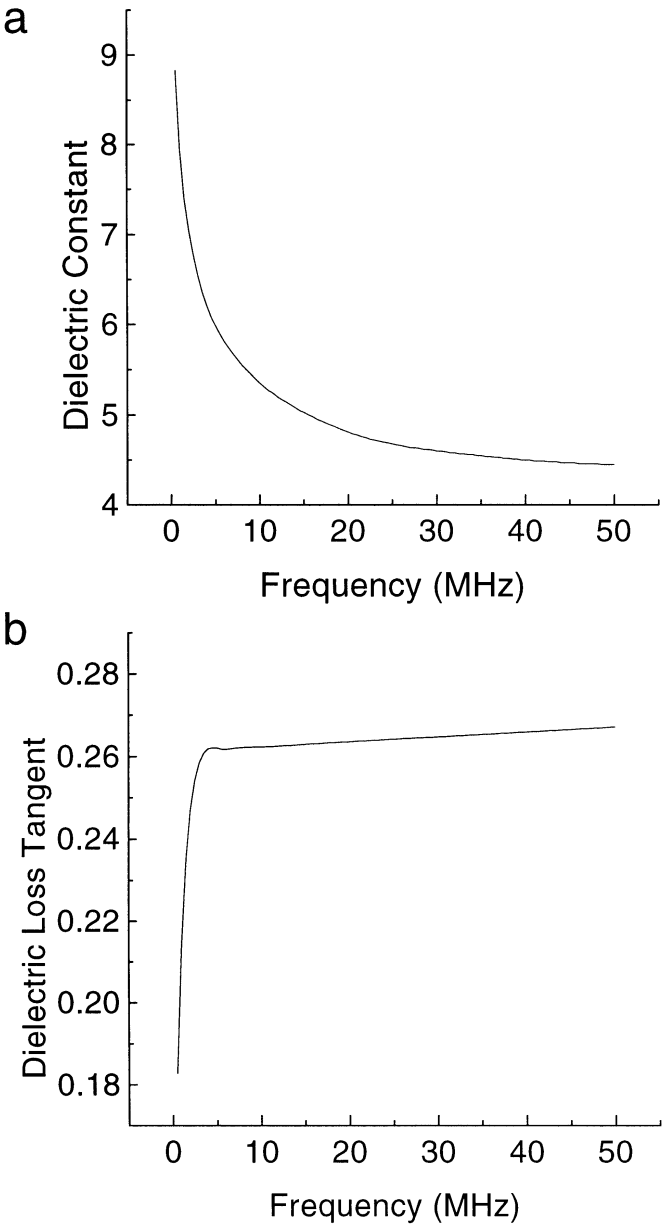

Fig. 8. Dielectric properties of PVDF over the frequency range from 0.5 to 50 MHz. Courtesy of Brown [17].

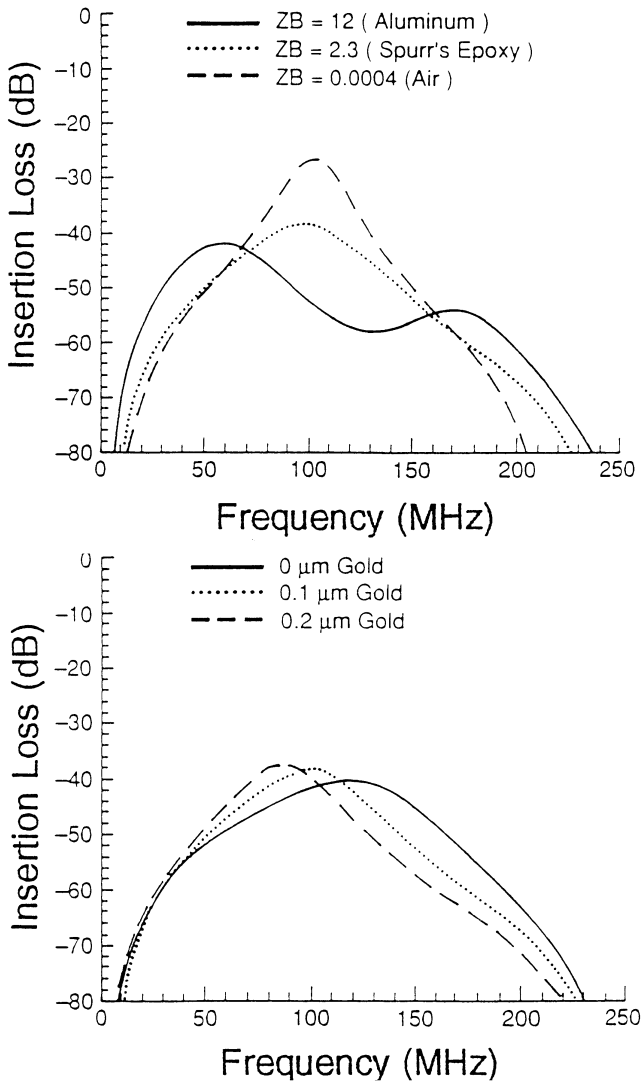

Fig. 9. Backing acoustical impedance (left) and electrode thickness (right) significantly impact electrical impedance of polymer transducers. Not only does the center frequency shift, but bandwidth is greatly affected. Courtesy of Sherar and Foster [18].

shows plots of the dielectric constant and dielectric loss tangent of PVDF up to a frequency of 50 MHz [17]. The relative dielectric permittivity drops from 9 to 4.5 over the frequency range from 1 to 50 MHz, and the dielectric loss tangent increases sharply at low frequencies and levels off at approximately 0.26 at frequencies above 5 MHz. Sherar and Foster [18] investigated the design and performance issues for high frequency polymer transducers in 1989. In that study, measurements of fundamental material properties were performed, and KLM modeling was used to predict the response of various high frequency transducer configurations. Impedance data for a number of PVDF transducer configurations are given in Fig. 9. Because of its low acoustical impedance, PVDF can resonate in either a $^1/_4$ or $^1/_2$ wave mode depending on the backing acoustical impedance. A 6-mm^2 area, air-backed, 9.8-μm thick PVDF layer resonates in the $^1/_2$ wave mode at 104 MHz; the same thickness of polymer resonates in the $^1/_4$ wave mode on an aluminum backing. The insertion loss plots of Fig. 9 (top) clearly show this behavior. Mass loading because of electrode thickness has a significant effect on the performance of polymer transducers. Fig. 9 (bottom) shows that as the electrode thickness increases from 0.0 to 200 nm of gold, the center frequency of the air-backed resonator drops from 120 to 85 MHz. Therefore, electrode thickness must be carefully controlled in the design of very high frequency probes. Our current PVDF transducer designs incorporate electrodes comprised of 30-nm chromium blended with 100 nm of gold. An exploded view of the parts for a 40- to 70-MHz PVDF/P(VDF-TrFE) transducer is given in Fig. 10. The important components consist of an SMA microwave connector (6 mm outer diameter), a plastic insert with inner diameter equal to the desired aperture

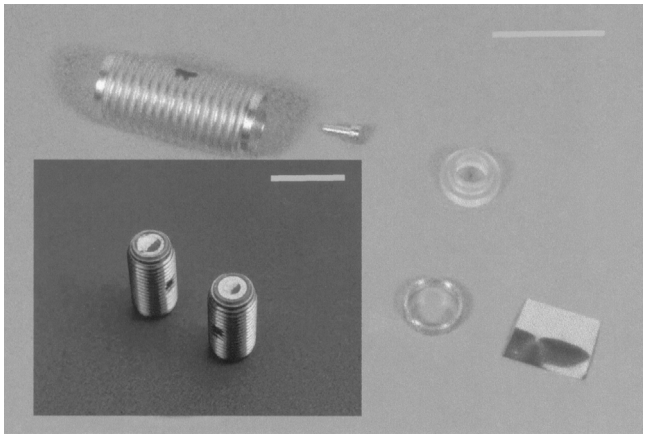

Fig. 10. Exploded view of the parts in a polymer/copolymer transducer. Inset shows two completed devices with different diameters and focal lengths. Bar = 10 mm.

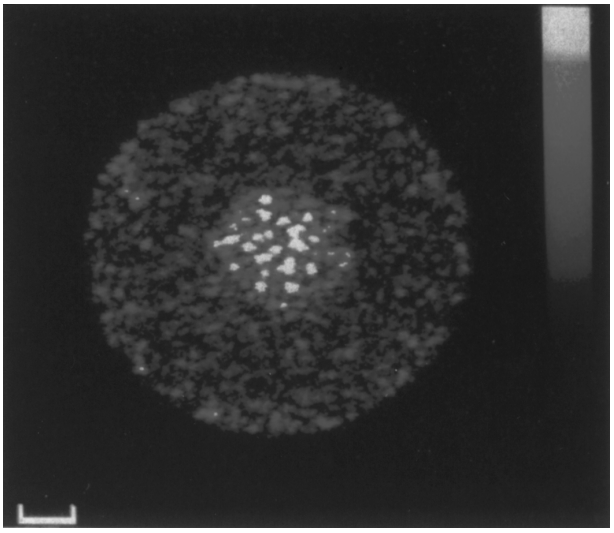

Fig. 12. Imaging of living tumor spheroids was reported in *Nature* by Sherar *et al.* [21] in 1987. A lightly backed 100-MHz PVDF transducer was used to make this striking image of a $\sim$ 800-μm tumor spheroid with a necrotic center. Bar = 100 μm.

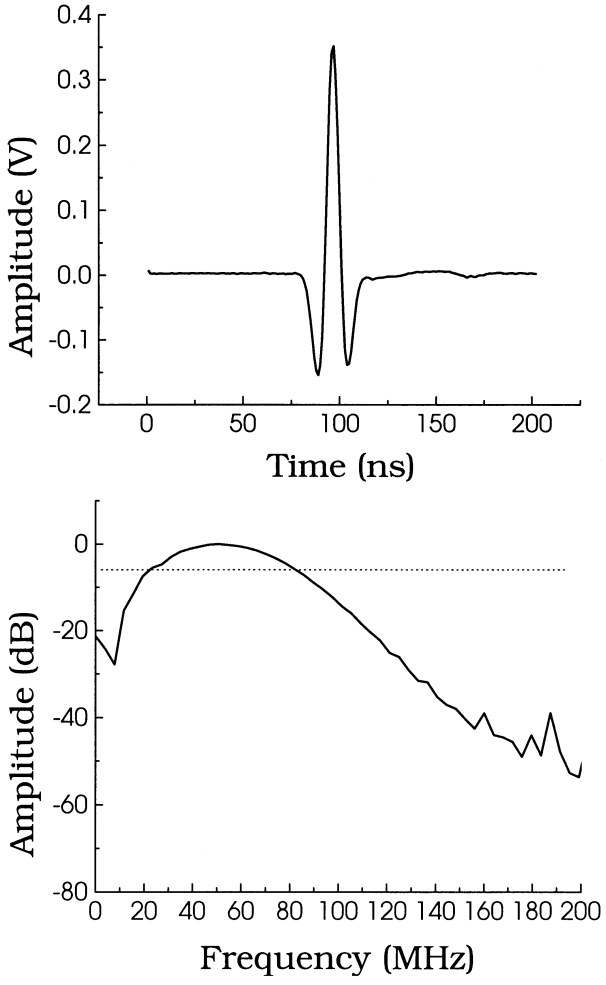

Fig. 11. The trademark of a good polymer/copolymer transducer is outstanding bandwidth as shown for this 4-mm diameter, 51-MHz copolymer transducer. Here, the echo from a reference flat demonstrates a 120% bandwidth.

size, a PVDF retaining ring to stretch the polymer across the aperture, and the previously electroded polymer sample itself. Not shown is the conductive epoxy backing and jig for spherically deforming the radiating aperture. The inset of Fig. 10 shows two completed devices with diameters of 2.5 and 3 mm and 6-mm focal lengths. Details on the fabrication of these devices are given in the work of Sherar and Foster [18], Lockwood *et al.* [19], and Foster *et al.* [20]. Polymer transducers made as described above work well in the 30- to 70-MHz range and provide bandwidth in excess of 100% and insertion losses of approximately 40 dB. An example of a received waveform from a plane quartz reflector at the focus of a 4-mm diameter P(VDF-TrFE) transducer similar to those shown in Fig. 10 is given in Fig. 11. The center frequency of this conductive epoxy-backed device is 51 MHz, and the bandwidth is 120%. The use of a low impedance backing doubles this center frequency for higher frequency operation [18].

IV. Medical and Biological Applications of High Frequency Polymer Transducers

The first high frequency images made with polymer transducers were reported by Sherar *et al.* [21]. These images were made of biological targets such as tumor spheroids in the C-mode format at 100 MHz. The benefit of imaging living tissue at microscopic resolution was immediately apparent in that the growth of the tissue could be studied noninvasively as a function of time. Striking images of the internal structure of spheroids revealed significant differences in backscatter from oxygenated cells at the periphery of the spheroid, hypoxic cells in the interior, and necrotic cells in the center. Furthermore, it was found that the actions of cytotoxic drugs could be monitored noninvasively on the basis of backscatter level [22]. Recently,

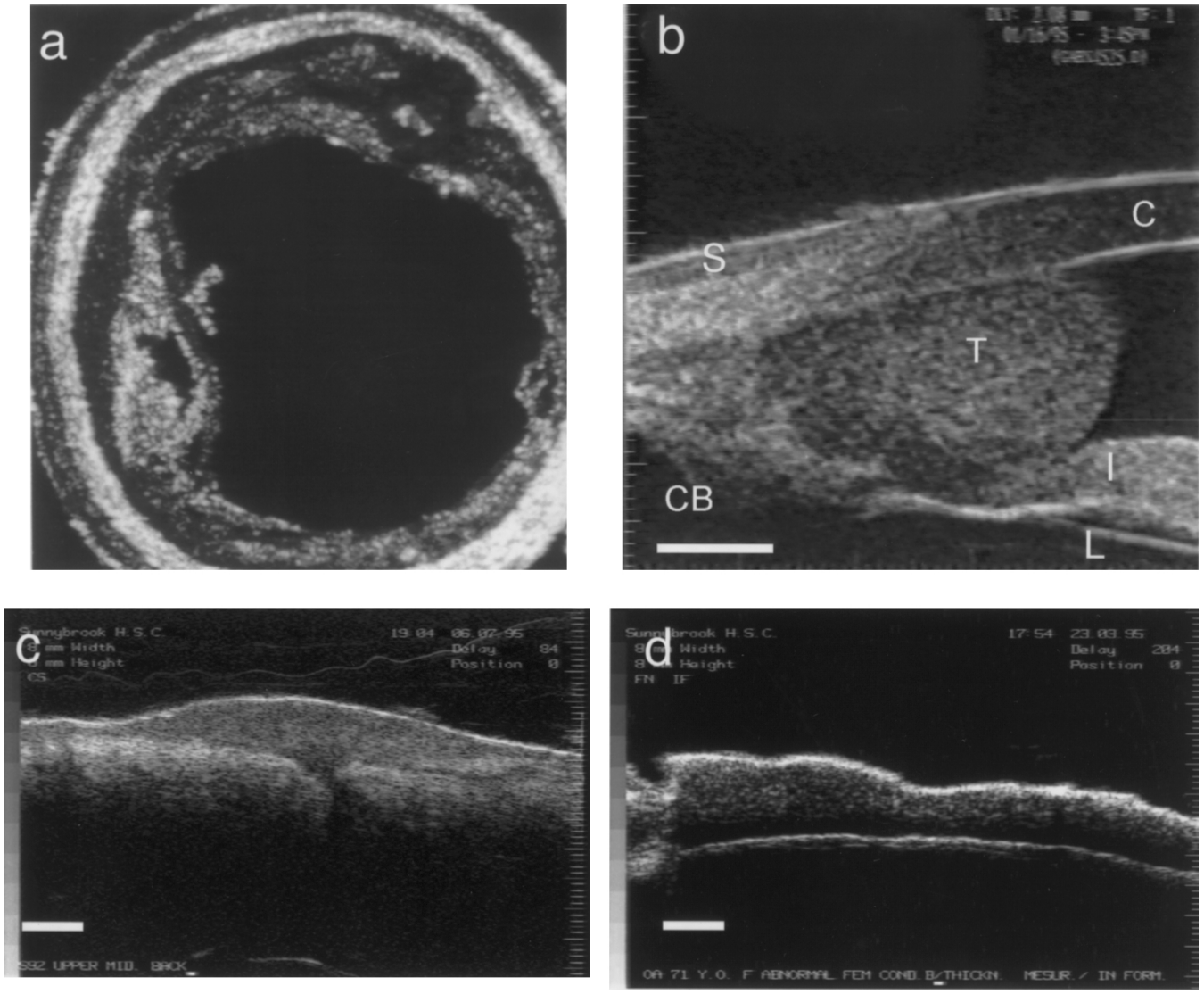

Fig. 13. UBM images of various human tissues: a) C-scan image plane taken perpendicular to the long axis of a diseased femoral artery ex vivo [25], b) in vivo UBM image of the anterior segment of a patient's eye with an ocular melanoma (C = cornea, CB = ciliary body, I = iris, L = lens, and T = tumor), c) in vivo UBM image of a superficial spreading melanoma, and d) ex vivo human osteoarthritic cartilage. Bar = 1.0 mm.

interest in this area has been rekindled with the discovery that cell apoptosis can also be differentiated by backscatter level [23]. An example of a spheroid image made using a 100-MHz low impedance backing PVDF transducer is given in Fig. 12. Here, limiting resolution is on the order of 15 μm. The penetration of the ultrasound allows visualization of subsurface planes unavailable to optical techniques. Success in simple biological systems, such as the spheroid, quickly focused attention on potential clinical B-mode applications. Ophthalmology applications centered in the 40- to 60-MHz range, and dermatology applications developed at lower frequencies of approximately 20 MHz. In addition to PVDF transducers, unique instrumentation was created to address the specific needs of high frequency applications. The ultrasonic visualization of living tissue at microscopic resolution is referred to in the literature as "ultrasound biomicroscopy" (UBM), "ultrasound backscatter microscopy," "high frequency ultrasound," or "very high frequency ultrasound". We use the term ultrasound biomicroscopy as it is analogous to optical biomicroscopy, a well-established optical method for visualizing living tissue in vivo. A recent review of ultrasound biomicroscopy is given by Foster *et al.* [24].

Fig. 13 shows examples of ultrasound biomicroscope images made with PVDF transducers of a wide range of human tissues. Fig. 13(a) represents a C-scan image plane taken perpendicular to the long axis of a diseased femoral artery ex vivo [25]. Outstanding definition of the complex wall structure is evident. From the outside, the layers visualized are the adventitia, media, thickened intima, and plaque. The internal structure of the plaque shows regions of lipid deposit and calcification. Images such as this are of great value in performing ultrasonic characterization studies such as those performed by [25]–[28]. The first clinical

applications of ultrasound biomicroscopy were developed in the field of ophthalmology by Pavlin and colleagues [29]–[32] and Sherar *et al.* [33]. Specialized instrumentation for corneal imaging has been developed by Silverman, Reinstein, and colleagues [34]–[36]. Recently, color flow imaging [37] of the eye has been reported by Ferrara *et al.* [37], [38]. As a result, commercial instrumentation is now available for routine clinical use of ultrasound biomicroscopy in ophthalmology. An example of an image of the anterior segment of a patient's eye with an ocular melanoma is given in Fig. 13(b). In this image, a tumor (T) is seen invading the ciliary body (CB) and iris (I). Also shown is the cornea (C) and lens surface (L). Several groups have developed high frequency skin imaging systems based on PVDF and copolymer transducers [39]–[42]. An example of an image of a superficial spreading melanoma taken from our own work is given in Fig. 13(c). This 6- x 8-mm image shows a weakly echogenic mass with a maximum depth of approximately 2 mm. The image of Fig. 13(d) shows a plane through osteoarthritic human cartilage. The cartilage and subchondral bone surface are clearly visualized. A more thorough discussion of the medical and biological applications of high frequency polymer transducers is given is given in [24].

V. Conclusions

PVDF and its copolymers have made substantial contributions as transducer materials for medical and biological ultrasound imaging. The unique features of these materials include low acoustical impedance, moderate coupling coefficients, rugged structure, and flexibility. Exploitation of these properties over the past 20 yr led, first, to the development and utilization of novel low frequency transducers and, subsequently, to the development of high performance high frequency transducers. In particular, the high frequency devices have led to a wealth of applications in the area of ultrasound biomicroscopy. Examples of these applications are amply depicted in this paper and have been extensively reviewed elsewhere [24]. Although more exotic materials such as relaxor ferroeletrics and composites show great promise for high frequency imaging, ferroelectric polymers such as PVDF and its copolymers will continue to dominate this field for years to come.

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