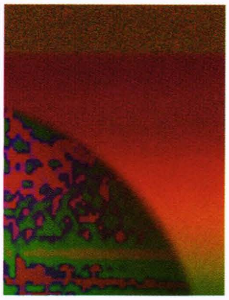

Aluminum Nitride on Sapphire Films for Surface Acoustic Wave Chemical Sensors

Joseph J. Suter, Wayne A. Bryden, Thomas J. Kistenmacher, and R. Daniel Porga

The development of a small surface acoustic wave (SAW) device on a piezoelectric aluminum nitride/sapphire substrate for possible use as a chemical vapor sensor is discussed. Aluminum nitride films on sapphire substrates were prepared, and prototype SAW devices with a resonant frequency of 267 MHz were designed and fabricated. Experimental results showed that the substrates support SAWs at the fundamental as well as the second and third overtone frequencies. This article presents the early results in the development of the piezoelectric aluminum nitride SAW chemical sensor. The design of the SAW device is described in detail, and initial results of its temperature and impedance characteristics are given.

INTRODUCTION

Commercially available vapor sensors and monitors are based on such diverse technologies as gas chromatography, gas-phase photoionization, interferometry, galvanic cells, solid-state conductivity cells, controlled combustion, infrared absorption, and mass spectrometry. The predominant technique for environmental gas monitoring in industrial and military settings is to draw a specific aliquot of gas through a tube containing an absorbing material. The sealed tube is sent to an analytical laboratory where the sample is released into a gas chromatograph or mass spectrometer. Although this method is accurate, it is costly, slow, and does not easily allow continuous workplace monitoring. To facilitate continuous monitoring, sensors based on some of the aforementioned technologies have been developed. Each sensor, however, has at least one of the following limitations: high cost, bulkiness, nonselectivity, or low sensitivity.

We describe in this article the design of a small surface acoustic wave (SAW) device for potential use as a continuous monitor of chemical vapors, particularly trichloroethylene (TCE). The design uses a piezoelectric aluminum nitride (AlN)/sapphire substrate that undergoes a change when exposed to TCE, which in turn causes a frequency change of the acoustic wave.

For many years, large quantities of chlorinated organic compounds have been used across a broad spectrum of industrial and military activities. TCE, perhaps the most prevalent of these substances, has been used as a solvent for degreasing and dry cleaning and has also been used in printing inks, paints, lacquers, varnishes, and adhesives. Recently, it has been found to be both carcinogenic and mutagenic in animal tests and hence is regulated by the Occupational Safety and Health Administration.

Despite the known occupational exposure hazards, TCE is still widely used instead of other less harmful solvents. Furthermore, much TCE-bearing waste is improperly disposed of, generally by dumping in industrial and commercial landfills. Chemicals such as TCE are known to leach out of the storage location and into the groundwater surrounding the landfill. The mobility of chemicals from a landfill into groundwater has caused such environmental disasters as the Love Canal incident in upstate New York. Therefore, early detection of TCE in the environment is critical. The SAW chemical sensor we describe in this article is inexpensive and designed specifically to measure small amounts of TCE in the environment. By measuring frequency shifts introduced into the SAW device through the interaction of the TCE and the AlN film, this device should be able to detect TCE vapors to levels smaller than 1 ppm.

SURFACE ACOUSTIC WAVE DEVICES

SAW devices make use of the piezoelectric effect exhibited by certain materials.[1] When an electric field is applied to a piezoelectric material, a mechanical strain is produced and vice versa. The relationship between electric field and strain is described by the coupling coefficient, which is a function of material and frequency. Two commonly used piezoelectric substrates are quartz and lithium niobate ($LiNbO_3$), with coupling coefficients of about 0.00116 and 0.056, respectively. Lately, AlN has been used as a piezoelectric material for the fabrication of SAW devices.

In SAW devices, an electric field is applied to the substrate by means of an interdigital transducer. In its simplest form, the transducer consists of many parallel metal electrodes called fingers, which are alternately connected together by bus bars as shown in Fig. 1. The fingers are usually a thin film of aluminum between 500 and 10,000 Å thick, many millimeters long, and a few micrometers wide; the spacing between fingers equals the width of the finger. Transducers are fabricated using photolithographic techniques developed for semiconductor technology.

When a time-varying voltage is applied to the bus bars, a spatially and temporally periodic electric field is generated within the transducer. This electric field in turn produces periodic strain fields in the substrate. Since quartz and $LiNbO_3$ are also elastic materials, acoustic (mechanical) waves result. This process is most efficient when the acoustic wavelength equals the finger pitch L; therefore, the finger width is related to the operating frequency of the device and the wave velocity

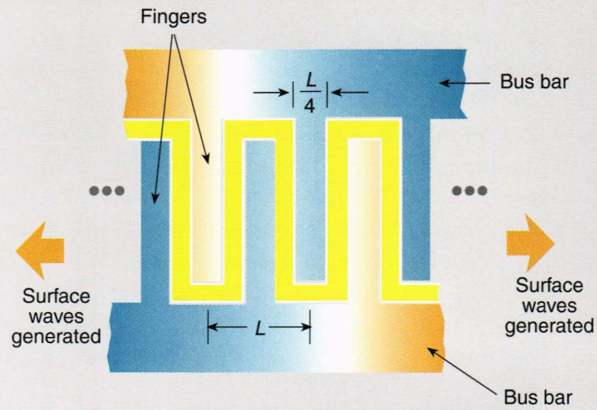

Figure 1. Schematic of the surface acoustic wave (SAW) device (resonance frequency = 267 MHz) placed on an APL-designed aluminum nitride substrate showing the interdigital transducers and SAW pattern (L = finger pitch in micrometers).

in the substrate material. For example, a 100-MHz device on quartz with a 3200-m/s wave velocity would result in fingers (3200 m/s)/(100 MHz)/4 = 8 μm wide.

Waves generated by the transducer propagate both at the substrate surface and into the bulk material; however, bulk waves are undesirable in a surface wave device and typically are scattered by roughening the bottom surface of the substrate. Because of the symmetrical nature of the transducer, surface waves are emitted from both apertures; hence, these transducers are called bidirectional. The waves in one direction are not needed and are absorbed by applying a lossy material on the surface. The waves in the other direction propagate nondispersively with velocities between 1,000 and 10,000 m/s and little attenuation. Owing to the piezoelectric effect, the traveling acoustic wave generates an electric field that can then be detected by a second interdigital transducer. The bidirectional nature of both the generating and detecting transducers will cause at minimum a 6-dB loss in the basic SAW device.

Since the detecting transducer is located a certain distance away from the generating transducer and acoustic waves are significantly slower than electromagnetic waves, a specified delay is introduced. This delay is useful in two ways: (1) it is the basis of the SAW delay line, and (2) it can be employed to separate output signals resulting from the acoustic wave from those generated by all other sources (feedthrough). Separation is accomplished by using a Fourier transform to look at the time response of the device. Since the acoustic response is separated in time from the feedthrough response, it is possible to remove the feedthrough response using an appropriate time window. Transforming the result back to the frequency domain gives a good picture of the acoustic response of the device.

By using more sophisticated transducer geometries, the device's response can be altered. One powerful technique, called apodization, involves varying the amount by which alternating fingers overlap. The voltage, produced as a surface wave excites each finger pair, is directly related to the amount those fingers overlap—the apodization strength. Thus, by controlling the apodization strength along the length of the transducer, a desired impulse response can be realized. Other techniques are also used and usually both transducers are modified; the device response, then, is the convolution of the individual transducer impulse responses.

The SAW device based on the AlN film described here is a filter centered around a fundamental frequency of 267 MHz and a bandwidth of 23 MHz. The frequency was selected on the basis of the availability of photolithographic masks as well as the large amount of information available on SAW devices at that frequency based on more conventional $LiNbO_3$ substrates.

CHEMICAL SENSING USING SURFACE ACOUSTIC WAVE DEVICES

Microfabricated SAW devices have been used for several years to measure gases and vapors.[2–5] The basis for these sensors is the modification of the SAW resonant frequency due to the condensation or absorption of gas-phase material onto the surface of the sensor. The fundamental equation determining the frequency shift Δf is

$$\Delta f = (k/a) f_0^2 \Delta m ,$$

where k is a material-dependent constant, a is the active area of the device, f_0 is the resonant frequency of the unloaded device, and Δm is the differential mass loading of the surface.

An examination of the equation reveals several design constraints on the application of SAW devices to chemical sensing. First, the frequency shift for a given mass loading is inversely proportional to the active area of the sensor and directly proportional to the square of the nominal operating frequency. In order, then, to achieve the highest sensitivity, the devices should be made small and operate at the highest frequency possible. Second, a severe constraint on the practical application of SAWs to environmental sensing is evident by the simple dependence of frequency shift on the mass of material deposited on the active surface. As given by the equation, the SAW device acts as a nonselective chemical balance responding equally to, for example, a microgram of water or TCE. A third design constraint is not evident in the equation but arises from a more exact treatment of SAW behavior. Generally, the operating frequency of a SAW device depends strongly on temperature. Complex temperature stabilization schemes often must be devised to minimize temperature-induced frequency shifts.[6]

The effects of liquid-phase acetone, isopropyl alcohol, and TCE on AlN ceramics were previously studied by Norton et al.[7] They observed that exposure of AlN to TCE resulted in a significant change in the material, whereas exposure to acetone and alcohol created very little change in the AlN films. Although further research into the exact nature of the TCE interaction with AlN is in order, these preliminary results indicate a strong specific interaction between TCE and AlN. It is this interaction that we exploited in the design of the AlN-on-sapphire SAW sensors. The mechanical, physical, and chemical properties of AlN make the SAW chemical sensors proposed here potentially superior to conventional devices in all aspects of the design criteria previously mentioned.

The SAW velocity for AlN (6000 m/s) is higher, by about a factor of 2, than any other material that has been used in the construction of SAW devices. In addition, AlN displays two other highly desirable properties: (1) a large electromechanical coupling constant and (2) very low dispersion at high frequencies. These factors combine to allow operation of AlN-based sensors at frequencies between 200 MHz and 1 GHz.

As noted earlier, the basic SAW sensor acts as a sensitive chemical balance without any selectivity to specific chemicals. A popular approach for achieving molecular selectivity with SAW devices entails the deposition of a chemically active coating on the piezoelectric surface.[7,8] The proper coating selectively adsorbs or reacts with the chemical of interest, producing a net change in mass loading and hence a change in the resonant frequency of the device and specific sensitivity to the molecule of interest. Given the highly desirable characteristics of AlN-based SAW devices, this approach is a viable option for sensing TCE. However, the realization of a truly specific, physically robust chemical coating on such sensors generally requires many years of research. For the sensors proposed here, a chemical interaction between the molecule of interest (TCE) and the piezoelectric coating (AlN) may have a more immediate application for environmental testing.

Historically, a significant drawback to the application of SAW devices has been the temperature dependence of the resonant frequency. Complicated schemes involving special crystal cuts and temperature stabilization systems have evolved to somewhat ameliorate this problem. For the work described here, the control of the acoustic properties by the precise control of piezoelectric film (AlN) thickness was exploited to develop nearly temperature-independent SAW devices. In fact, for frequency control applications (for example, resonators, delay lines, and filters), temperature-independent SAWs were fabricated.[5,9–11] These studies showed that by controlling the product $\mathbf{k}H$, where $\mathbf{k}$ is the wave vector ($2\pi/\lambda$) of the SAW and H is the AlN film thickness, devices could easily be produced with a nearly temperature-independent frequency near 1 GHz. For AlN films deposited on the basal plane of sapphire, a value of $\mathbf{k}H = 3.75$ gave the desired temperature independence near room temperature.[11]

ALUMINUM NITRIDE ON SAPPHIRE SURFACE ACOUSTIC WAVE DEVICES

For many years, single crystals have dominated the piezoelectric materials field, particularly quartz for bulk wave devices and $LiNbO_3$ for SAW devices.[10–12] However, many applications require piezoelectric materials in thin film form. High-quality piezoelectric thin films can perform well. In addition, they can be produced cheaply and are applicable to selective area coatings.

Of particular interest here are thin films of AlN, a Group IIIA nitride compound that is inherently piezoelectric with several desirable physical properties. As stated before, the SAW device, when optimally prepared, has a nearly temperature-independent operating frequency.[5,9,11] Furthermore, AlN-based SAW devices can operate at frequencies in the gigahertz range—a decisive advantage when developing sensor systems.[10]

The AlN films were deposited using a technique that relies on the creation of a magnetically confined plasma close to a target material. The positive ions from the plasma are accelerated to the target, where

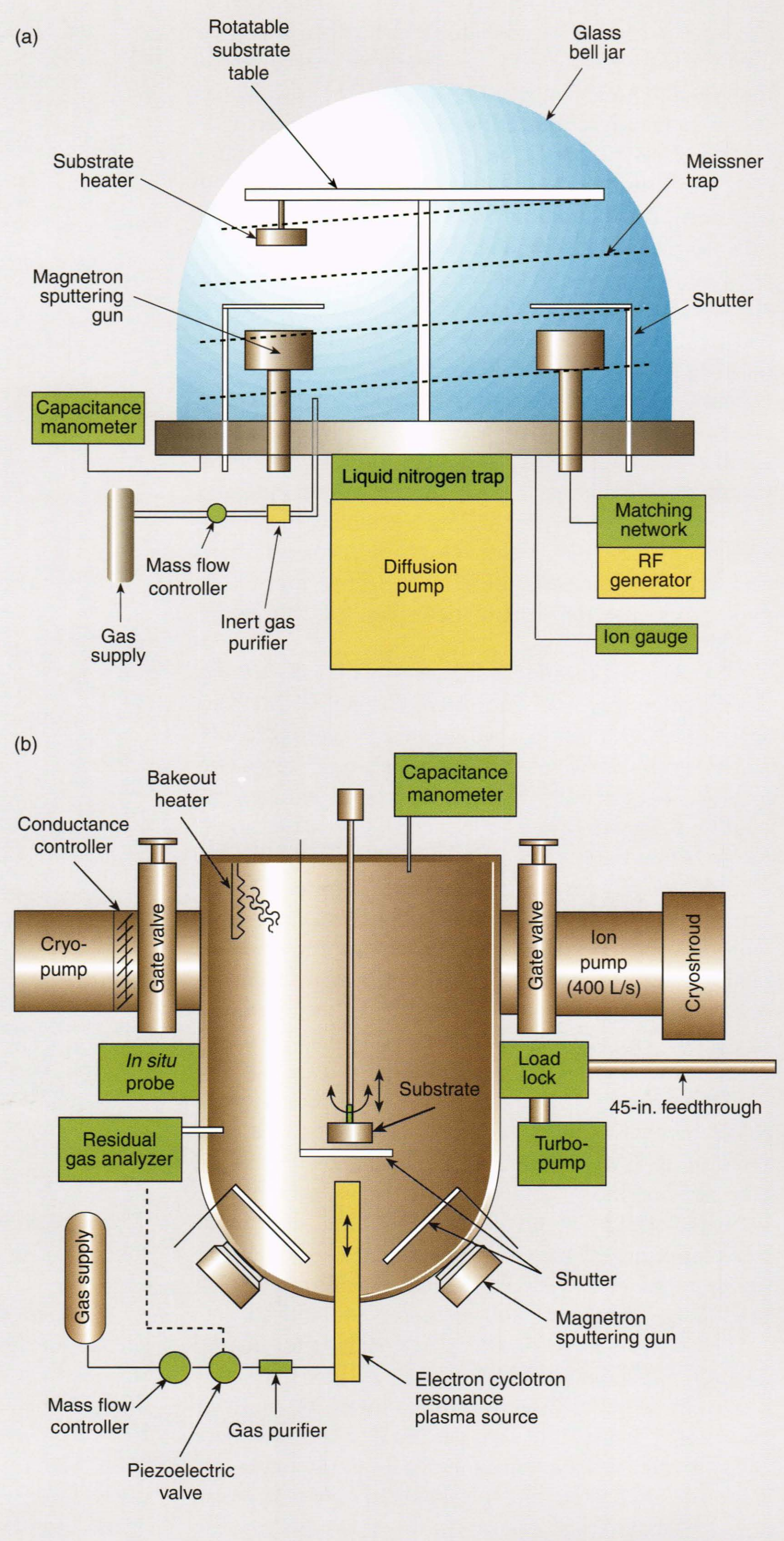

Figure 2. Sputtering systems used to deposit AlN films onto sapphire substrates: (a) high vacuum magnetron sputtering system, and (b) ultrahigh vacuum electron cyclotron resonance–assisted reactive magnetron sputtering system.

they eject the target material into the vapor phase, allowing it to coat a substrate held nearby. We have used this approach at APL for several years to produce high-quality piezoelectric samples of AlN.[13] Two automated magnetron sputtering systems have been employed. One system is based on a liquid nitrogen–trapped diffusion-pumped vacuum station capable of pressures on the order of 5×10^{-8} Torr (Fig. 2a), the other on ultrahigh vacuum technology with a base pressure of 8×10^{-11} Torr (Fig. 2b) and an additional electron cyclotron resonance (ECR) source of reactive nitrogen species. The latter system is particularly useful for this study because ultrahigh vacuum conditions minimize sample contamination, and the ECR source ensures chemical stoichiometry in the films. AlN prepared on heated single-crystal sapphire substrates using the sputtering approach is highly crystalline, optically flat, and chemically inert and has extremely high electrical resistivity (10^{12} Ω-cm). These physical properties, coupled with its outstanding piezoelectric properties, make this material highly desirable for the SAW sensors.

We produced several batches of AlN films during the development process. The direct-current magnetron-sputtered AlN films on sapphire were processed as substrates for the SAW sensors. The sapphire substrate had a thickness of about 500 μm. All samples were examined by X rays to assure proper crystallographic orientation before the devices were placed on the substrates. The AlN layers showed good structural properties, although samples of this thickness (1–1.5 μm) are somewhat beyond the coherence length for the highest-quality sputtered growth. X-ray diffractometer scans for some samples, grown at 600 and 900°C, are displayed in Fig. 3a. The (00.2) peak at 36° is sufficiently narrow to indicate good crystal quality, as shown in Fig. 3b.

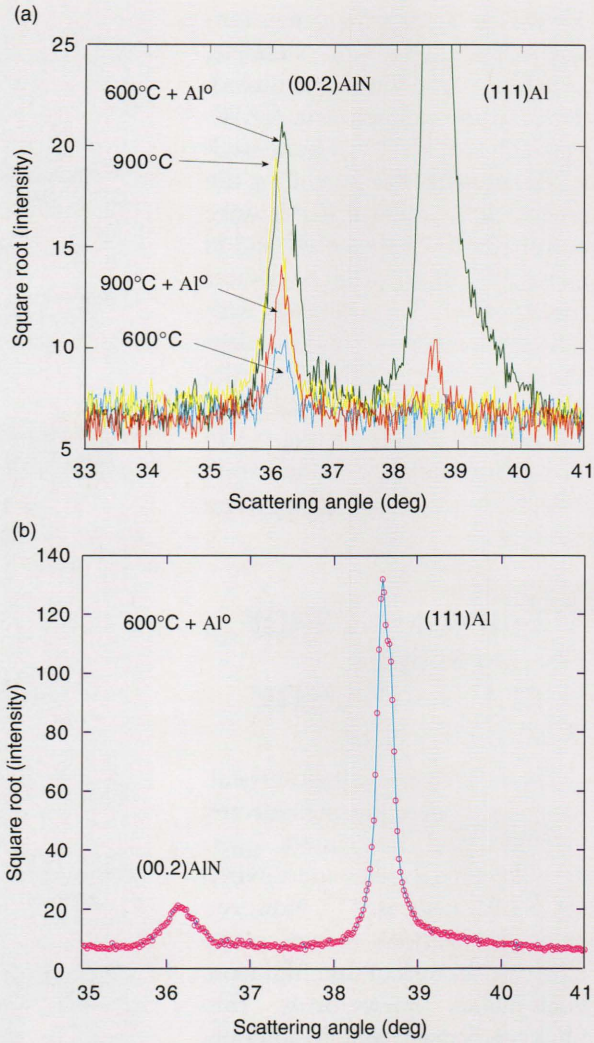

Figure 3. X-ray scattering results from AlN/sapphire substrate. (a) Responses at 600 and 900°C capped with elemental aluminum (Al°). (b) Total counts for (00.2)AlN and (111)Al (crystallographic orientation). The AlN film thickness was about 1.5 μm and was deposited on a 500-μm sapphire substrate.

After manufacturing the AlN substrates, several SAW bandpass filters (originally designed to be centered at 184 MHz with 18 MHz of bandwidth when fabricated on $LiNbO_3$) were developed and subsequently characterized. Six devices were built and their input and output impedances matched. Figures 4a and 4b are photographs of the SAW device. Scanning electron microscope images are shown in Figs. 4c and 4d.

The devices were tuned to 184 MHz (the original $LiNbO_3$ SAW device frequency), and their frequency responses were measured using a network analyzer and signal generator. No passband responses were initially observed, but periodic ripples in the frequency responses indicated the presence of some acoustic signal. A fast Fourier transform was performed on the data to observe these impulse responses, and a signal 0.55 ms after the feedthrough peak was observed. Knowing the delay (0.7995 μs) as well as the wave velocity (3975 m/s) of the $LiNbO_3$ device, the wave velocities of the sample AlN/sapphire devices were calculated: (3975 m/s)(0.7995 μs)/(0.55 μs) = 5778 m/s. This approximates theoretical and experimental results (about 5700 m/s) obtained previously by Tsubouchi et al.[11]

We next calculated the expected center frequency of the sample devices. Since the same transducer geometry was used on the AlN samples as on previous $LiNbO_3$ devices, the acoustic wavelength must be the same for both. The center frequency of the AlN devices is thus (184 MHz)(5778 m/s)/(3975 m/s) = 267 MHz. The devices were then tuned to this frequency and measured again. Signal processing was performed on each device's frequency response to remove the feedthrough signal, resulting in a frequency response due to the acoustic signal alone. The results of all samples clearly showed a characteristic passband response centered around 267 MHz with 23 MHz of bandwidth (Fig. 5). The

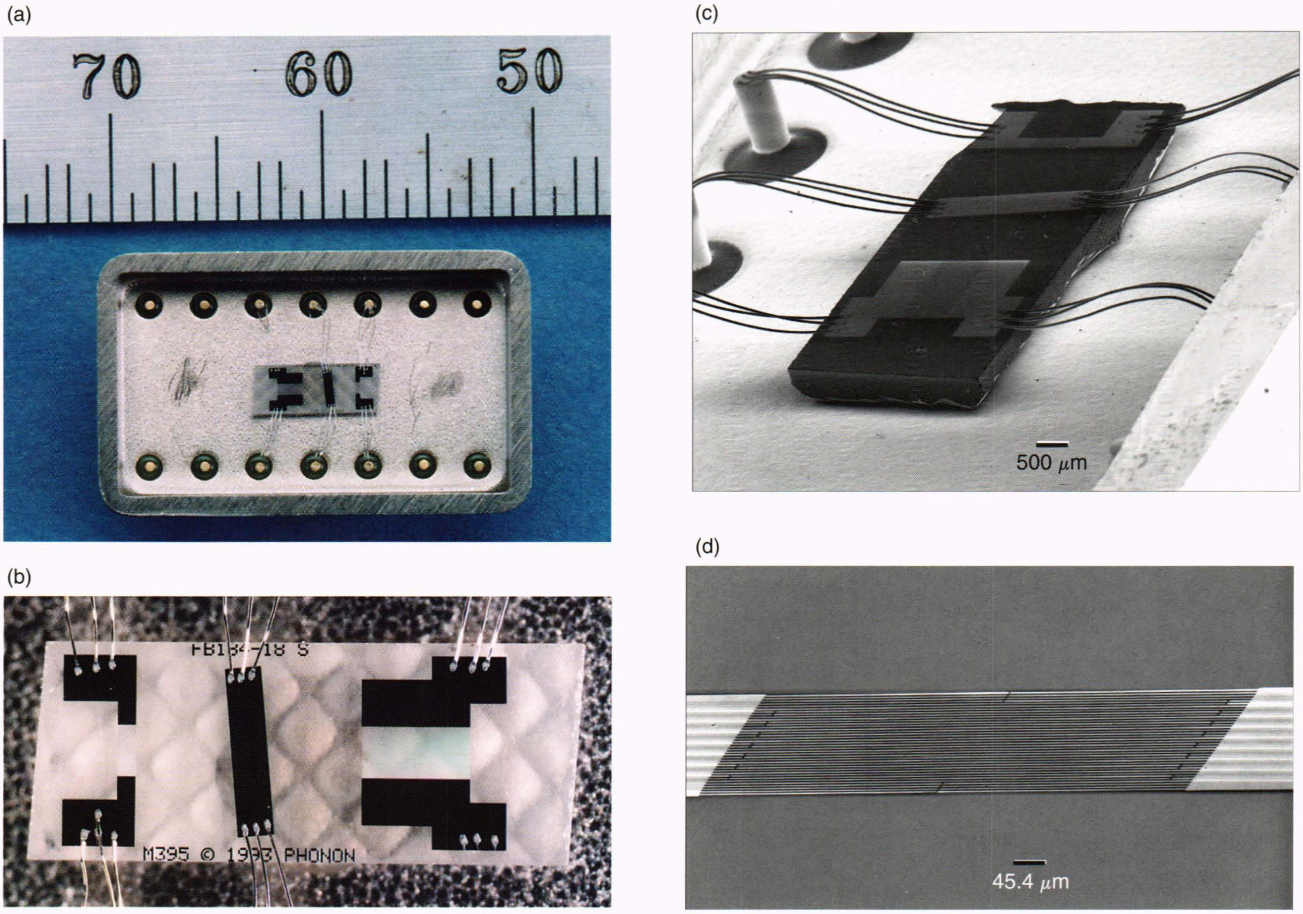

Figure 4. The 267-MHz AlN/sapphire SAW device: (a) package layout, (b) detail with interdigital transducers, (c) scanning electron microscope (SEM) image (2 kV) at a magnification of 20, and (d) SEM detail of the SAW structure at a magnification of 220.

center frequency of these devices also verified that the phase velocity was close to its actual value.

The response of the SAW filters was also characterized by measuring their impedance. Figure 6 shows that the impedance is about 22 Ω, given the present impedance matching network.

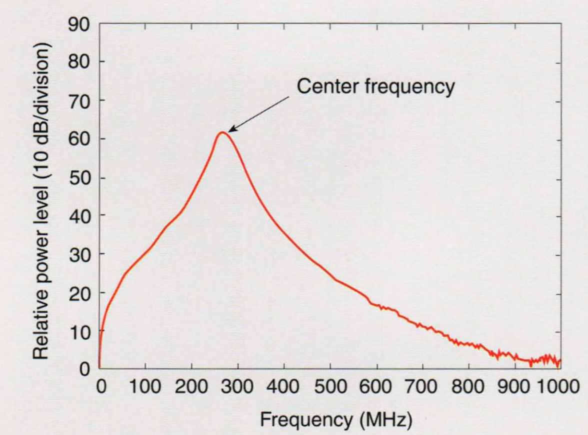

Figure 5. Frequency response of the SAW device (center frequency of filter = 267 MHz).

These results indicate that the AlN/sapphire SAW devices were functioning properly, aside from having a higher than expected insertion loss. This loss can be attributed to the value of the coupling coefficient as calculated by Tsubouchi et al.[11] Again, if **k** is the wave vector and H the thickness of the AlN film (the AlN film on our samples was approximately 1–1.5 μm thick), then $\mathbf{k}H = 2\pi/[(5778 \text{ m/s}) / (267 \text{ MHz})](1\ \mu\text{m}) = 0.290$. This value corresponds to a coupling coefficient of about 0.00025. The coupling coefficient of LiNbO3 is typically about 0.056, more than 200 times greater than that measured for the AlN/sapphire SAW devices. However, the coupling coefficient can be dramatically increased by using an AlN film with greater thickness and/or by increasing the device's operating frequency.

CONCLUSIONS

Initial results of the development of a SAW chemical sensor on AlN/sapphire substrates are promising. Through signal processing we determined that SAWs

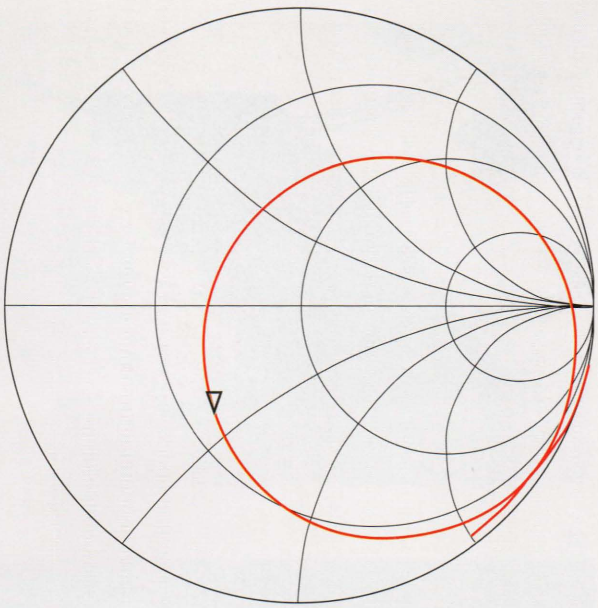

Figure 6. Smith chart of the SAW filter (impedance ≈ 22 Ω). Marker shows response at 267-MHz AlN on sapphire.

were successfully generated and detected on these substrates. However, higher than expected insertion losses occurred, which were attributed to the thickness of the AlN film and the frequency of operation of the SAW device. We are now implementing a design modification for the next generation of sensors by increasing the AlN thickness and designing them with a resonant frequency of about 900 MHz. Given the successful design of the 267-MHz AlN/sapphire SAW device, measurements on the interaction of TCE vapor with the AlN films can proceed. Concurrently, improvements to the electrical coupling in the SAW sensor can be addressed. Future development of this sensor will also focus on making the AlN film selective to compounds other than TCE.

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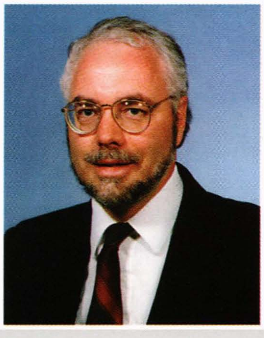
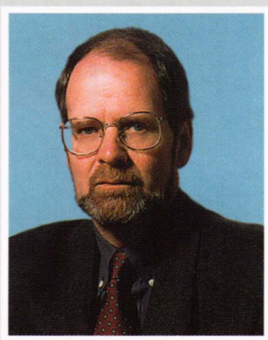
THE AUTHORS



JOSEPH J. SUTER received a B.S. degree in physics and mathematics from the Free University of Amsterdam, The Netherlands, in 1977. He received an M.S. degree in physics from Michigan State University in 1980 and an M.S.E.E. degree from the University of Maryland in 1983. In 1988, he was awarded a Ph.D. degree in materials science and engineering from The Johns Hopkins University. Dr. Suter joined APL in 1983 and is a Principal Professional Staff scientist and Supervisor of the Space Department's Time and Frequency Section. He is a member of the IEEE, APS, SPIE, and Sigma Xi. He has served on several IEEE committees on time and frequency technology. Dr. Suter was appointed a research associate in the Department of Materials Science and Engineering of The Johns Hopkins University in 1993. He is the (co)author of over 50 technical publications. His e-mail address is Joseph.Suter@jhuapl.edu.

WAYNE A. BRYDEN is a chemist in the Sensor Science Group of the APL Research Center. He obtained a B.S. degree in chemistry from Frostburg State University in 1977, and M.S. and Ph.D. degrees in physical chemistry from The Johns Hopkins University in 1982 and 1983, respectively. He conducted graduate research at APL from 1978 to 1982 and was employed as an APL postdoctoral fellow in 1982. In 1983, he joined APL as a Senior Staff Chemist. His current research interests include materials physics, mass spectrometry, magnetic resonance, miniaturized sensor technology, and chemical and biological detection. He is a member of the American Chemical Society, the American Physical Society, the American Vacuum Society, the Materials Research Society, and Sigma Xi. Dr. Bryden is listed in *American Men and Women of Science* and is the author of over 60 scientific publications. His e-mail address is Wayne.Bryden@jhuapl.edu.



THOMAS J. KISTENMACHER is a Principal Professional Staff chemist in the APL Research Center. He obtained a B.S. degree in chemistry from Iowa State University and M.S. and Ph.D. degrees, also in chemistry, from the University of Illinois. During 1969–71, he was a junior fellow in the A. A. Noyes Laboratory for Chemical Physics at the California Institute of Technology. From 1971 to 1982, he served on the faculties of The Johns Hopkins University, Homewood, and the California Institute of Technology. Dr. Kistenmacher joined APL in 1982 as a member of the Microwave Physics Group. He subsequently served in the Materials Science Group and currently is a member of the Sensor Science Group. His principal research interests include the fabrication and analysis of thin films for a variety of structural, electronic, and optical applications, X-ray diffraction, and crystalline structure and structure–property relationships in a variety of electronic and magnetic materials. His e-mail address is Thomas.Kistenmacher@jhuapl.edu.

R. DANIEL PORGA received a B.S. degree in electrical engineering from the University of Connecticut in 1993. Since then he has worked as an engineer at Phonon Corporation, a manufacturer of SAW devices. Mr. Porga is a member of the IEEE and the Tau Beta Pi honor society.