Informacije MIDEM 24(1994)4, Ljubljana UDK 621.3:(53+54+621+66), ISSN0352-9045

FERROELECTRIC THIN FILMS FOR APPLICATIONS IN MICROELECTRONICS AND IN MICROMECHANICS

Nava Setter
Laboratoire de céramique, EPFL
Ecublens, Lausanne, Switzerland

INVITED PAPER
22nd International Conference on Microelectronics, MIEL'94
30th Symposium on Devices and Materials, SD'94
September 28. - September 30., 1994, Rogla, Slovenia

Keywords: microelectronics, micromechanics, thin films, ferroelectric films, practical applications, new applications, recent developments, new products, FRAM, PZT ferroelectric materials, DRAM memories, high dielectric constants, piezoelectric micro actuators, pyoelectric sensors, IR image sensors, IR imaging

Abstract: Because of their unique properties, ferroelectic thin films are of interest for non-volatile memories, for DRAM's, for applications as integrated sensors (e.g. IR sensors) and for a variety of micro mechanical devices such as micro motors and micro pumps. An overview of recent developments is presented here, with emphasis on fabrication related issues and on properties relevant to the above applications.

Feroelektrične tanke plasti in njihova uporaba v mikroelektroniki in mikromehaniki

Ključne besede: mikroelektronika, mikromehanika, plasti tanke, plasti feroelektrične, aplikacije praktične, aplikacije nove, razvoj nedavni, proizvodi novi, FRAM feroelektrični RAM, PZT materiali feroelektrični, DRAM pomnilniki, konstante dielektrične visoke, mikroaktuatorji piezoelektrični, senzorji piroelektrični, IR senzorji slik, IR upodabljanje

Povzetek: Zaradi svojih edinstvenih lastnosti lahko feroelektrične tanke plasti uporabimo pri izdelavi nebrisnih pomnilnikov, DRAM pomnilnikov, integriranih senzorjev (npr. IR senzorjev) in za izdelavo raznovrstnih mikromehaničnih naprav, kot so to mikro motorji in mikro črpalke.
V prispevku podajam pregled rezultatov najnovejših raziskav in dosežkov s poudarkom na izdelavi in lastnostih izdelanih struktur z zgoraj naštetih področij uporabe.

INTRODUCTION

Whereas bulk ferroelectrics are in use since long time, high quality ferroelectric thin films have been fabricated only recently. This new development allows for the first time the integration of ferroelectrics with silicon and opens the way to the realisation of a variety of new products, the first of which (a pyroelectric detector) has just appeared on the market /1/. Among the potential applications are ferroelectric non-volatile memories, DRAM storage capacitors and various micro-electromechanical devices. It is the large microelectronics companies who are interested in the memory applications of ferroelectrics, and the development is accelerating in particular in Japan. As for the applications in micromechanics, the interest is growing in medium size industries and is strong in particular in Germany and in Switzerland. In the latter field, the applications are just beginning to be discovered.

FERROELECTRIC RANDOM ACCESS MEMORY (FRAM)

Ferroelectric materials, such as Pb(Zr,Ti)O_3 (PZT), possess a permanent dipole that can be reversed with electric field (Fig. 1). Being such, they make a non-volatile memory. When thin ferroelectric films are integrated with CMOS circuits, they can form a solid-state non volatile memory. The properties of the ferroelectric films are ideal for memories: Read and write is very fast (nano seconds) /2/, the needed voltage for operation is small (3-5 V) and the stored information is maintained for long duration (high retention, no refresh needed). Other advantages are its potentially very high storage density and its insensitivity to radiation. The films show potential

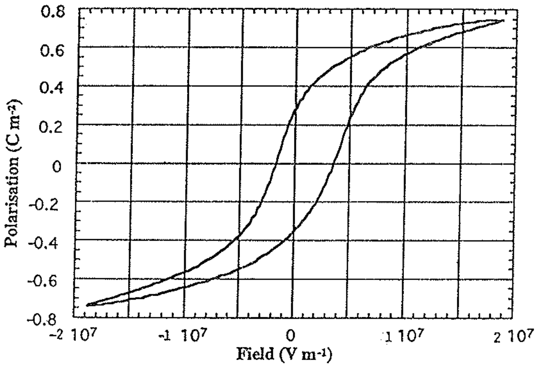

Fig. 1: The ferroelectric hysteresis loop.

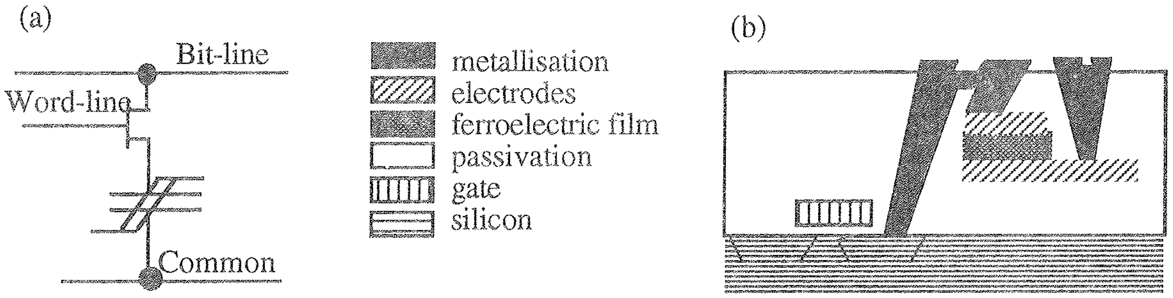

Fig. 2: *Description of a ferroelectric non-volatile memory. a) schema, b) cross section.*

to be compatible with VLSI processes and therefore the memory could have the same performance as that of DRAM with the advantage of being non volatile. Fig. 2a and 2b show one of the current proposed structure of the ferroelectric memory.

One of the important characteristics of a memory is the difference between the switched and the non switched signal after a pulse is given. Fig. 3 shows a typical behaviour of a "well prepared" PZT film on platinum metal electrode. The decrease in the signal, referred to as "fatigue", constituted a major problem of the films. Two solutions have been proposed: a) the use of a ceramic conductor such as RuO_2 instead of platinum which is reported to eliminate the fatigue problem at least up to 10^{13} cycles /3/, b) the use of an undisclosed material which is claimed to be fatigue free and has been patented /4/ and is used by Matshushita and Sony for the memory development.

The fabrication of the ferroelectric layers is still in development. The material mostly studied until now is PZT, showing a high spontaneous polarisation, a low coercive field and a high resistivity. The three methods suitable for a large scale fabrication are sol-gel technique, sputtering and MOCVD. Presently MOCVD, with its high growth rate and good step coverage is gaining importance /5/. The electrodes have a strong influence on the performance of the memory cell. The bottom electrode has to withstand the high processing temperature of the film (600 -700 °C) and the corrosive atmosphere (oxygen and lead vapours). Problems of lead migration into the silicon, through the electrodes, and problems of diffusion of Si or Ti (Ti is used as adhesion layer between the SiO_2 and the platinum) have been detected. The upper electrode is as important. Its deposition is done after the ferroelectric layer is processed, but problems are encountered related to adhesion and to the possible existence of a passive layer or a gap between the upper electrode and the PZT film (Fig. 4). The processing and the integration problems are being studied presently at numerous industries. Recently, researchers at Philips have shown reliable operation of memory cells (25 μm^2 area) of PZT with Pt electrodes, at 3V supply voltage and 20 n s pulse-width, up to 10^{13} cycles /6/.

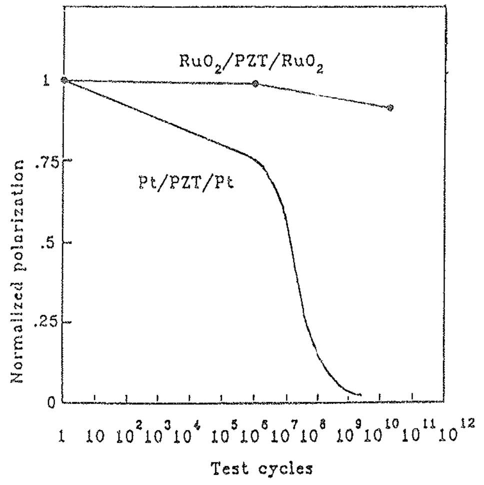

Fig. 3: *Schematic curve comparing fatigue of films with Pt electrodes and with RuO_2 electrodes.*

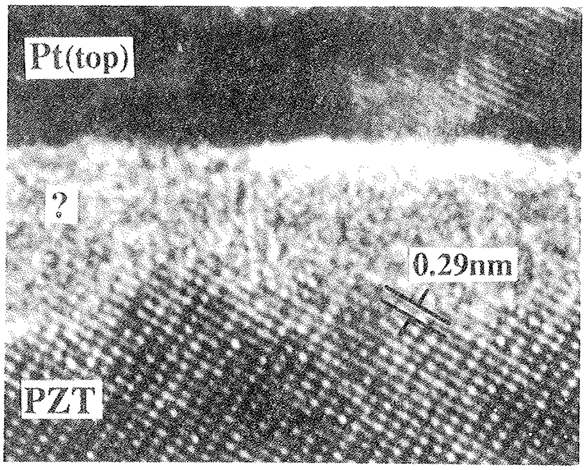

Fig. 4: *TEM micrograph of PZT film with Pt upper electrode showing a gap between the film and the electrode (courtesy of I.Reaney, Laboratoire de céramique, EPFL)*

HIGH DIELECTRIC CONSTANT MATERIALS FOR DRAMS

In parallel to the development of ferroelectric non-volatile memories, there is a current effort to replace SiO_2 capacitors of dynamic random access memories with very high dielectric constant films. The reason for this is the need to increase DRAM density beyond that achievable by low permittivity dielectrics. Until now the increase in density has been obtained by a reduction in the capacitor area and a subsequent reduction in the capacitor thickness, but low thickness limitations have been attained and an increase in density beyond 1Gbit is hardly foreseeable even with Ta_2O_5 whose dielectric constant is higher than that of silica (ε_r = 17 and 4 respectively). Ferroelectrics with high permittivity would seem advantageous, since they allow reduction in the capacitor area while maintaining a reasonable thickness. However, the paraelectric $(Ba,Sr)TiO_3$ (BST) seems more appropriate for this application, since it possesses high permittivity up to the high frequencies envisioned for ULSI DRAMs (GHz regime) and its losses at these frequencies are much lower than those of the high permittivity ferroelectrics. Low leakage current (<1.5fA at Vcap (3V)) and null time dependent dielectric breakdown (TDDB) at Vcap for long duration (10 years) /7/ are the most important impositions on the new capacitor material. Recently Koyama et al. /8/ and others have shown large charge storage capabilities of BST and Taguchi /9/ has shown that this material performed better than thin Si_3N_4 with regards to leakage current. Activity in this area is pursued in most DRAM producing industries, in particular in Japan /10/.

PIEZOELECTRIC MICRO ACTUATORS

The application of piezoelectric films in micromechanics is presently at the demonstration level. Piezoelectric micromotors based on ZnO thin films have been fabricated /11/, and more recently a PZT micromotor has been demonstrated with rotational velocities 100-200 rpm and torques in the pN-m/V^2 range /12/. ZnO has the advantage of ease of deposition and low permittivity while ferroelectric ceramics, like PZT, have much larger piezoelectric coefficients. Until now, the piezoelectric properties of the ferroelectric films have hardly been investigated and the question whether the piezoelectric properties of the films are similar to those of bulk ceramics is still open. The answer to this question is needed before commercialisation can take place. Direct piezoelectric measurements (measurements of the induced piezoelectric charge under alternating force) on poled $Pb(Zr_{0.53}Ti_{0.47})O_3$ films of $\approx$ 1 μm thickness showed piezoelectric coefficient $d_{33} \approx 130$ pC/N/13/. This result is not sensitive to the ac pressure excerted (Fig. 5a) but is less than half of the bulk value. The converse piezoelectric effect is studied using optical interferometry and the results are consistent with those obtained by the direct method. Fig. 5b shows a typical piezoelectric hysteresis loop of PZT obtained by laser interferometry. The piezoelectric coefficient is dependent on the applied DC bias and, unlike the case of bulk ceramics, is significantly reduced upon removal of the bias. The origin of the effect may be due to clamping of domain walls either at the electrode-film interface or in the bulk. A clear answer is still missing. The piezoelectric response of a micromachined cantilever beam has been measured as well /14/. In this configuration, precise determination of the coupling coefficient k_{13} (the efficiency of conversion from electrical to mechanical energy) can be done. 0.4 μm thick films have shown k_{13} = 0.15, or about half of the value reported for bulk PZT ceramics. DC bias was necessary in order to obtain this coupling coefficient.

Few demonstrations have been presented by now concerning potential applications. A new configuration of a micromotor has been presented, with enhanced coupling between the piezoelectric (ZnO) stator and the metallic rotor. This motor includes standard micromechanical components with components similar to those used by the watch industry /15/. The same configuration has also been used recently with PZT /16/. Another

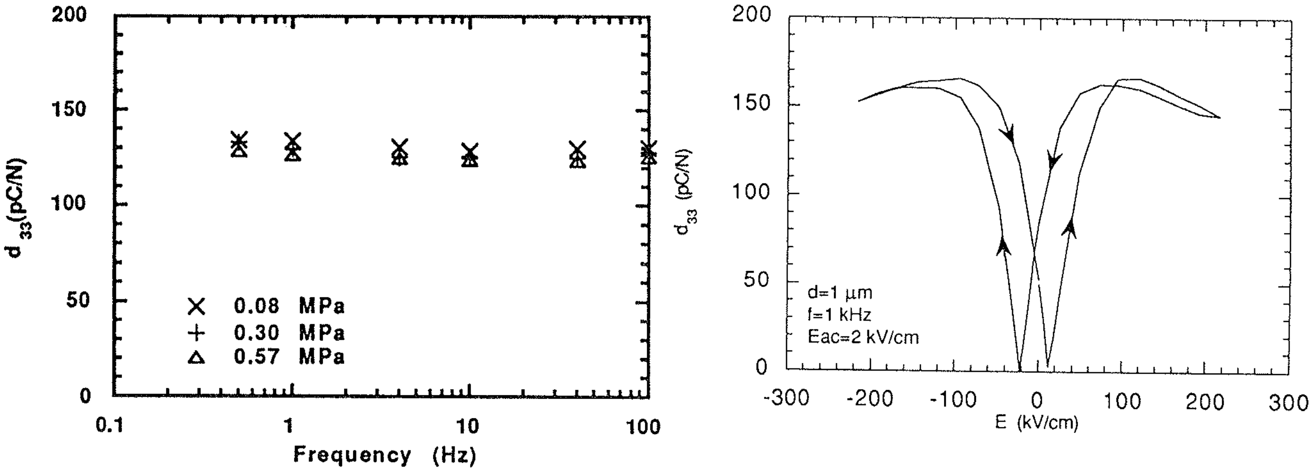

Fig. 5: *Piezoelectric properties of PZT films. a) d_{33} as a function of ac force frequency measured in the direct method, b) Piezoelectric hysteresis loop of a 1.0μm thick film, measured by interferometry (courtesy of A. Kholkine and D. Damjanovic, Laboratoire de céramique, EPFL).*

direction which has been pursued first with ZnO and then with PZT /14/ is the development of a piezoelectric micropump where travelling waves carry the liquid forward. One of the potential application of such pumps is in continuous drug delivery. Another possibility is the use of a tubular micropump in a structure similar to that shown in Fig. 6a. In this case, a microtube with outer diameter 30 μm and wall thickness 5 μm has been fabricated from ZnO and coated with Pt electrodes /17/. A longitudinal wave could then generate the pumping action. Piezoelectric micro-beams on micro-machined silicon have been proposed for sensors and actuators. Fig. 6b shows such a structure.

Piezoelectric coatings on optical fibers have been used to create an optical phase modulator. The piezoelectric coating would deform under the applied field, stressing the fiber and modulating the optical signal. A family of interferometric sensors is expected to result from this work /18/.

For piezoelectric applications, thin films thicker than 1 micron are needed. Processing studies are being carried out presently in order to develop high quality "thick" thin films. The problem is the control of stoichiometry during the long period of heat treatment necessary for the growth process. Fig. 7 shows PZT films of thickness 1μm. In the films prepared by the sol-gel methods (Fig. 7a) a lead deficient layer is seen at periodicity of 0.3 μm, resulting from the thermal history of the film. The sputtered films (Fig. 7b) are highly crystalline, but demand a higher thermal budget for the preparation.

PYROELECTRIC SENSORS

Generating charge under temperature variations, pyroelectrics crystals and bulk ceramics are often used as infra-red sensors. When the pyroelectric elements are fabricated in a matrix array form, they allow the extraction of spatial temperature distribution and its temporal variation and make efficient IR imaging systems.

The fabrication of pyroelectric thin films adds the two following advantages: The pyroelectric signal, being inversely proportional to the element thickness, is enhanced by the thickness reduction (up to a certain thickness the level of which is dependant on the configuration of the component), and therefore thin films have potential for better performance than the bulk pyroelectrics. Secondly, the possibility to deposit the pyroelectric film on silicon has the potential to allow the integration of the device with the needed electronics.

A suitable material for pyroelectric applications is lead titanate, $PbTiO_3$, doped with lanthanum. Lead titanate is useful due to its high pyroelectric coefficient ($p = 180$ $\mu cm^{-2}K^{-1}$) and low dielectric constant ($\varepsilon_r = 180$). The lantanum is known to further enhance the pyroelectric coefficient. The preparation of bulk lead titanate ceramic is difficult because of cracking that occurs due to the large distortion at the phase transition. Lead titanate thin films do not suffer from this problem. In addition it is possible to prepare these films on MgO substrates with their c-axis perpendicular to the surface so that the figure of merit is optimised /19/.

In order to optimise the pyroelectric sensor performance, low thermal conductivity between the film and the substrate is needed. In this case, micromachining technology is of great use. Ye et al. /20/ have deposited the active element on a sacrificial layer which was later removed, leaving the active element above a cavity and supported by a polysilicon membrane (Fig. 8a). In this way, the high thermal conductivity of Si does not degrade the device performance. Another possibility to avoid the heat conductivity by the silicon is to etch the silicon below the active element, leaving only a thin supporting membrane /21/ (Fig. 8b). Weda et al. /22/ have shown

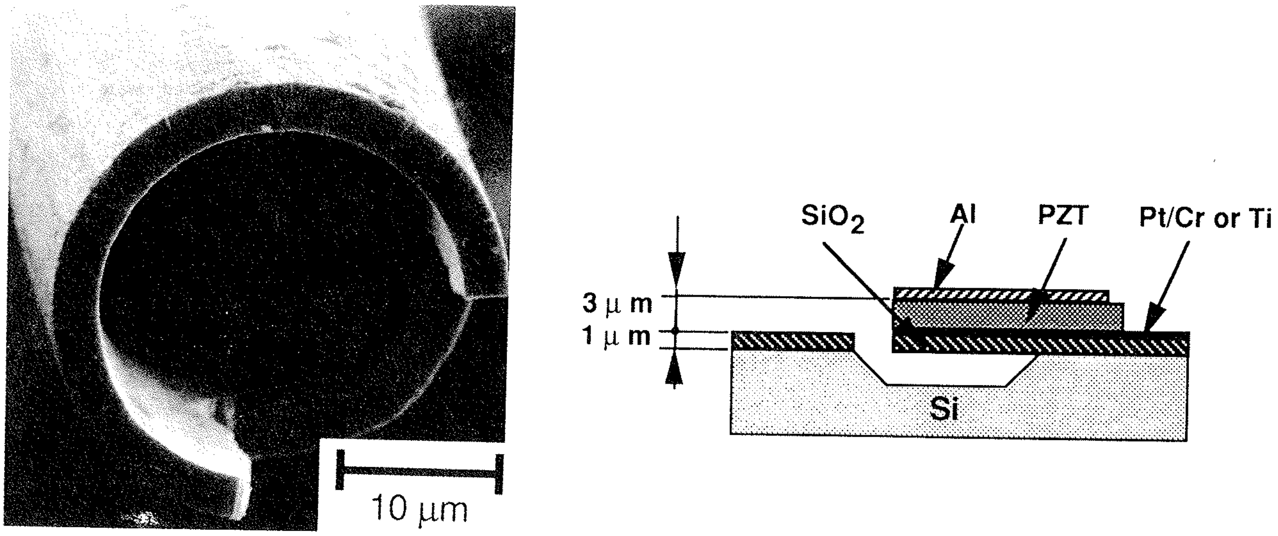

Fig. 6: Various piezoelectric microcomponents. a) piezoelectric microtube and b) schematic view of a piezoelectric micro beam (courtesy of G. Fox and K. Brooks, Laboratoire de céramique, EPFL).

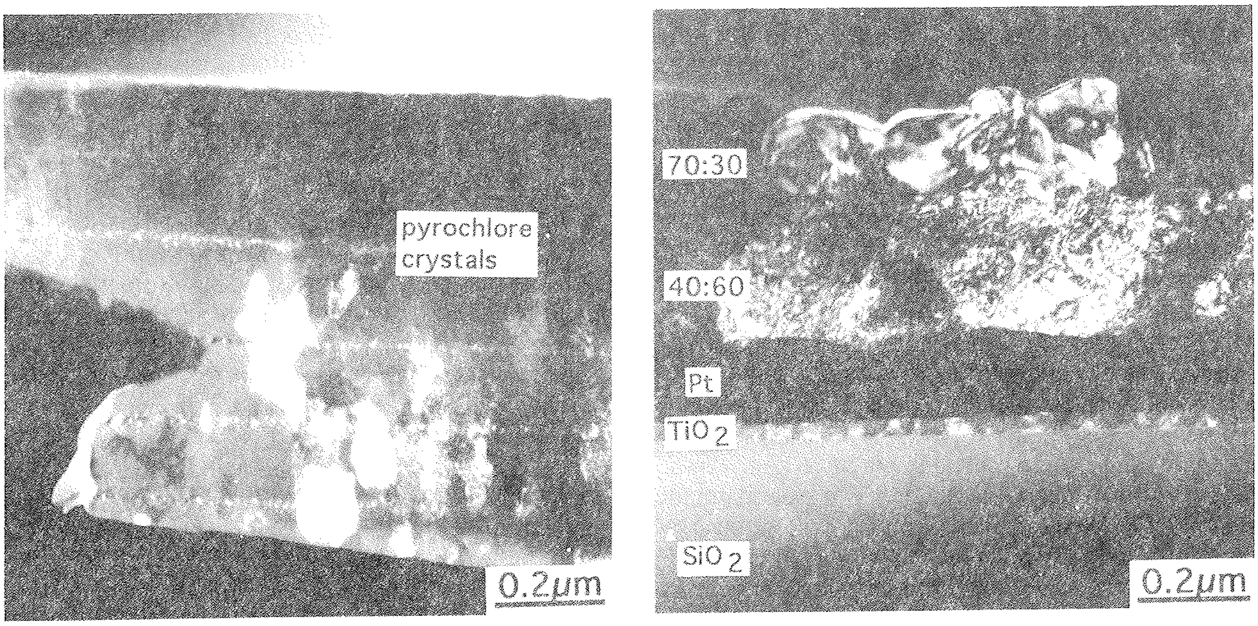

Fig. 7: Microstructure of 1 micron PZT film: a) sol gel film , b) sputtered film.

another efficient configuration. Lead titanate film was grown on MgO to provide preferred orientation growth with optimised properties. Then a support, thermal insulating layer, has been grown on the active element and the MgO has been etched away. The structure was then bonded to a supporting ceramic substrate (Fig. 8c).

FINAL REMARKS

The current experiments related to the performance of FE films indicate numerous advantages for their use in many applications. At the same time it is clear that the addition of the ferroelectric layer on standard Si devices, whether in microelectronics or for micro sensors and actuators, means additional fabrication costs. It is not clear yet whether the additional advantages will be attractive enough to allow for the extra cost. In the meantime, development is proceeding in both industry and academic laboratories throughout the world.

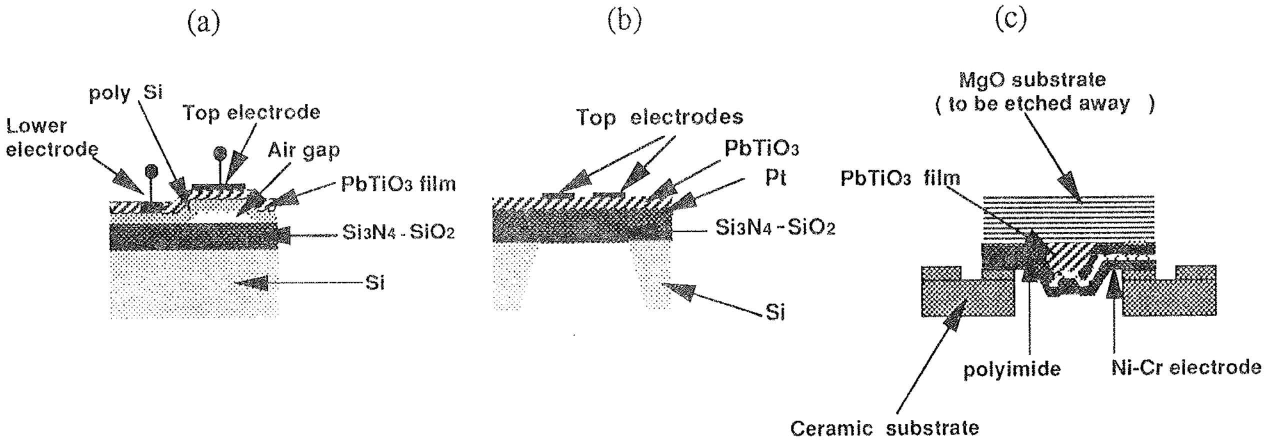

Fig. 8: Proposed configuration for pyroelectric micro-sensors. a) sacrificial layer method /20/, b) micromachined silicon method /21/, c) inverted MgO method /22/.

ACKNOWLEDGEMENT

The EPF Council is acknowledged for financial support (Materials Priority Program).

REFERENCES

/1/ K. Iijima, Presented at the MRS Spring Meeting, San Francisco, USA, 1993

/2/ P. Larsen et al., Appl. Phys. Lett. 59, 611 (1991)

/3/ I.K. Yoo and S.B. Desu, Phys. Stat. Sol (a), 133, 565 (1992)

/4/ J.F. Scott et al., presented at ISIF'92, ISIF'93, ISIF'94

/5/ Ferroelectric News, 3,2, Summer 1994

/6/ P. Larsen, presented at COST-514 Workshop on Ferroelectrics, Stockholm, May 1994

/7/ P. Larsen, G. Spierings, R. Cuppens and G. Dormans, Microelectronic Eng. 22, 53 (1993)

/8/ K. Koyama et. al., IDEM Tech. Dig. 823 (1991)

/9/ M. Taguchi, IEEE Elec. Dev. Lett. 13, 642 (1992)

/10/ T. Shiosaki, presented at COST-514 Workshop on Ferroelectrics, Stockholm, May 1994

/11/ R.M. Moroney, R.M. White and R.T. Howe, Proc. IEEE Ultrason. Symp. 1989, 745

/12/ A M. Flynn, L.S. Tavrow, S.F. Bart, R.A. Brooks, D.F. Ehrlich, U.K. Udayakumar and L.E. Cross, Proc. IEEE Ultrason. Symp. 1990, 1163

/13/ K. Brooks, D. Damjanovic, A. Kholkine, I. Reaney, N. Setter, P. Luginbuhl, G.-A. Racine, N. De Rooij and A. Saaman, Integrated Ferroelectrics, 1994, in press

/14/ K.B. Brooks, D. Damjanovic, N. Setter, Ph. Luginbuhl, G.-A. Racine and N.F. de Rooij, Integrated Ferroelectrics, in press (1994)

/15/ G.-A. Racine, R. Luthier and N. F. De Rooij, Proc. IEEE MEMS-93 Workshop, 1993, 128

/16/ P. Muralt, T. Maeder, M. Kohli and N. Setter, Proceedings of the 1st Swiss Conference on Materials for Engineering Systems, Sion, Switzerland, September 1994

/17/ G.R. Fox and P. Danai, J. Mater. Research, in press

/18/ G.R. Fox, M. Kosec, P. Danai and N. Setter, presented at th Electroceramics IV Conference, Aachen, Germany, September 5-7, 1994, Silicate Industries, in press

/19/ K. Ijima, R. Takayama, Y. Tomita and I. Ueda, J. Appl. Phys. 60, 1986, 2914

/20/ C. Ye, T. Tamagawa, P. Schiller and D.L. Polla, Sensors and Actuators A 35, 1992, 77

/21/ A.J. Bell, Y. Huang, O. Paul, Y. Nemirovsky and N. Setter, Integrated Ferroelectrics, 1994, in press

/22/ Weda et al., reported in ref. 10 above

prof.dr. Nava Setter
Ecole Polytechnique Fédérale de Lausanne
Laboratoire de céramique, EPFL
Ecublens, Lausanne, CH-1015 Switzerland
tel.: 021-693 29 61, fax.: 021-693 29 35

Prispelo (Arrived): 30.10.1994 Sprejeto (Accepted): 22.11.1994