Ultrasonic PZT gas sensor using thick film technology

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Abstract

A new low cost ultrasonic gas sensor for human environment pollution monitoring was developed using a thick film piezoelectric PZT ceramic resonator. The working principle of this kind of sensor is based on ultrasonic resonance of gas flowing through a cell. A PZT piezoelectric ceramic resonator generates ultrasonic waves within the cell cavity. Stationary waves are generated if distance between a vibrating surface and the opposite parallel reflecting wall, is an integer multiple of half vibration wavelength of the compound oscillator, formed by the vibrating surface and surrounding gas. In this way, resonance is induced in the gas and it absorbs an important amount of vibration energy, so changes can be measured on oscillation amplitude.

Sensors based on this principle were designed using quartz crystals as ultrasound generators. Nevertheless in resonance condition the quality factor of quartz is much better than of PZT ceramic used in this case, several advantages are remarkable: low cost due to thick film technology, much higher tolerance to temperature variations and the possibility of PZT ceramic fine mechanical adjusting. The resonating cell's wall could be adjusted by using PZT elements as piezoelectric actuators, which control the cavity gap.

Introduction

In recent years interest in gas detection has been increasing. A reliable gas sensor should be sensitive, selective and stable.

Different classes of gas sensors are now available. In particular different ultrasonic techniques are used. One of these techniques detects the change of ultrasonic phase wave transmitted through the gas when the gas composition changes [1]. Mecea et al. [2,3] proposed an ultrasonic resonance method using a quartz resonator as a transducer based on the Energy Transfer Model [4]. It states that when a gaseous medium is in contact with the vibrating surface of a piezoelectric resonator, a compound resonator is formed and ultrasonic longitudinal waves will be propagated in the surrounding gas, taking into account that the resonator is vibrating in thickness mode.

Gas is compelled to resonate at the same frequency $\mathbf{f_q}$ of the piezoelectric resonator. When the resonance condition is accomplished standing waves are generated between the surface of the quartz resonator and the surface of a parallel reflecting wall. In this situation the gas absorbs an important amount of vibration energy. The resonance condition is:

$$l = n\,V/2 = \; n\,v_g/2\,f_q$$

This kind of sensor must have a good temperature control of the detection cell because the values of the resonance parameter **l** and the ultrasound velocity $\mathbf{v_g}$ in the gas are strongly affected by temperature.

Based on this method, present work deals with the development of tunable gas sensor in thick film PZT piezoelectric technology. We try to take advantage of the ceramics stability with temperature to build the cell cavity and the ultrasonic resonator using the same technology. It is shown also how this technique helps us to improve parallelism between cell roof and PZT upper surface.

However since the resonator is strongly dumped, is difficult for the electronic circuit to keep up oscillations, because it has to provide all the energy. This is a problem to be solved. In particular this fact is more important when the piezoelectric materials used are less electrical-inductive than quartz.

Another difficulty arises for PZT resonators to keep resonating conditions because of its low **Q** when compared with quartz, and it perturbs frequency stability. This fact is due to the important presence of other resonance modes besides the thickness mode (e.g. radial mode).

In this paper the thick film technology used to obtain thick piezoelectric ceramics and the cell cavity design are described. In order to obtain a sensitive resonator, electrical impedance characteristics of the quartz, piezoceramics piezocomposites 1-3 [5] and thick film piezoceramics are analyzed.

Thick film sensor technology

A thick film sensor circuit is normally considered to be one that comprises layers of special inks deposited onto an insulating substrate. The method of film deposition is screen-printing using a finely woven mesh of stainless steel mounted under tension on a metal frame [6]. The mesh is coated with an UV sensitive emulsion onto which the sensor pattern can be formed photographically. The ink is placed on the screen and a squeegee traverses the screen under pressure, forcing the ink through the open areas of the mesh. The next stage of the process is to dry the film removing organic solvents from the paste in a conventional box oven. The dried film is relatively immune to smudging and the substrates can be handled.

The films themselves contain fine powders which must be exposed to high temperature in order to form a solid, composite material. This is called ‘sintering’, and is achieved in a belt furnace.

Thick Film Ink

Any paste for electronic thick film passive component or sensing element contains at least three ingredients: an organic vehicle which gives the correct paste rheology, an active ingredient which gives its electric and sensing properties and a glass frit or an oxide, which is the bonding agent to the substrate.

The vehicle normally consists of ethyl cellulose or other cellulose derivatives, dissolved in a blend of solvents. We have used for this purpose a commercial component, the vehicle 403 from ESL, dissolved in terpineol as main ingredient and xylene to adjust viscosity while printing.

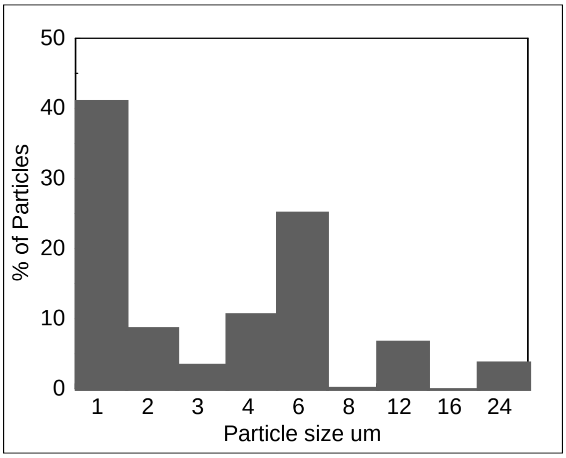

Fig. 1

The active ingredient is a milled powder from PZT5 piezoelectric discs that were previously sintered. Particle size affects characteristics like sinterability and density, so discs were milled in a tungsten carbide colloidal type mill to a particle mean size of few micrometers and then sieved with a 510-mesh screen. Particle size distribution of the final powder can be seen in Fig. 1.

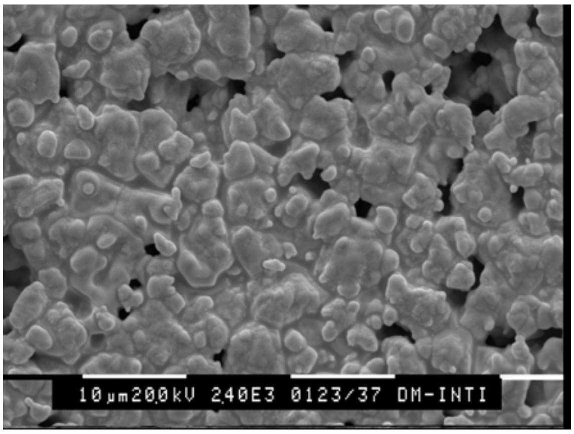

Fig. 2

The final powder has a 5% in weight of a glass frit with 600°C softening point and 2% of LiF to improve sintering properties.

Printing process was done with a 250 mesh, and dried at 150°C during 30 min. Sintering temperature was 900°C and it was kept for 20 min.; whole cycle lasted 90 min. Figure 2 shows the surface of the sintered sample.

Thick Film Cell

The main component of the cell is the PZT element, a disc of about 10 mm diameter and 200 μm thick over an alumina substrate. It was built by four layers of PZT, printing sequentially and sintered separately. The PZT upper surface was ground before printing the electrode.

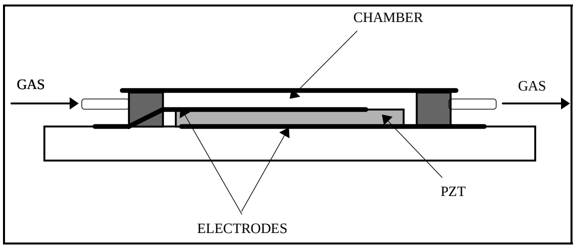

Fig. 3

To perform lateral walls of the cell we used two ceramic annular rings, with 22 mm inner diameter and approximately 100 μm thick each one. These elements were glued each other and to the substrate with a high temperature (850°C) thick film glass, allowing us to obtain a very thick wall with few layers in one step. The cell base is the substrate and the roof is a 316 stainless steel foil.

Standing waves can be obtained if the distance between the upper surface of PZT and the opposite parallel wall is an integral number of half wavelengths of the ultrasound generated by the resonator. The gap between the PZT and the roof can be mechanically adjusted to any length close to the resonance condition.

Another design for lateral walls was made with three PZT stacks, ring sectors-shaped at 120°. Stack connections were made to obtain maximum displacement with minimum voltage. This configuration helps to improve parallelism between cell roof and PZT upper surface, applying different DC voltage to each stack. In all cases Heraeus C4303 conductor ink was used.

Last step is to polarize the PZT. Different approaches were attempted, using electrical fields from $2{\cdot}10^6$ to $3{\cdot}10^6$ V/m during 15 to 120 min. Also different temperatures were used to optimize the poling process. Best results were found with an electric field of $2.8{\cdot}10^6$ V/m for 40 min. at 150°C.

Measurement gas system

For characterization of gas measurement sensors, a very reproducible and reliable system is needed to produce a gas mixture with controlled concentration, temperature and humidity. The system has fine adjust flow velocity of the gas mixture, also using velocities below 1 mm/s, and keeps constant flow while the mixture is changing. Use of toxic or combustible gases is allowed.

Gas sensor characterization systems look for reproduce ambient conditions where the devices are going to work. Most of the time it means a low gas velocity but sometimes knowledge of sensor step response is needed, so we choose an intermediate solution. Using a sensor chamber of 25 ml volume we can get a gas concentration pulse width of about 1 minute, for flow velocities of 1 mm/s or higher.

Sometimes we need to produce a very low gas flow, so the manifold must be mounted in a horizontal plane to avoid problems with the hydrostatic pressure. On the other hand, some types of sensors require high gas concentration in the carrier. For this reason the system has three flow control lines of 2000, 500 and 5 ml/min. To keep constant the gas flow when switching different gas concentrations, an electronic valve system switches the gas flow to the sensor or vents it without stopping the flow.

Impedance analysis

Oscillator circuit used to make the ceramic resonator is a standard Pierce configuration. Oscillation condition of such a feedback oscillator is that the loop-gain must be greater or equal than unity and total phase on the loop must be zero [7,8]. The piezoelectric element used as resonator is placed in the feedback network and must have a large reactance-frequency slope on the oscillation range, i.e. its impedance must change so rapidly in this range that all other frequency dependent circuit elements can be considered to be of constant reactance.

Next figure shows the impedance and phase characteristics of an X-cut quartz crystal (fig.4.) and a 100% ceramic PZT disc (fig. 5) around the resonance frequency.

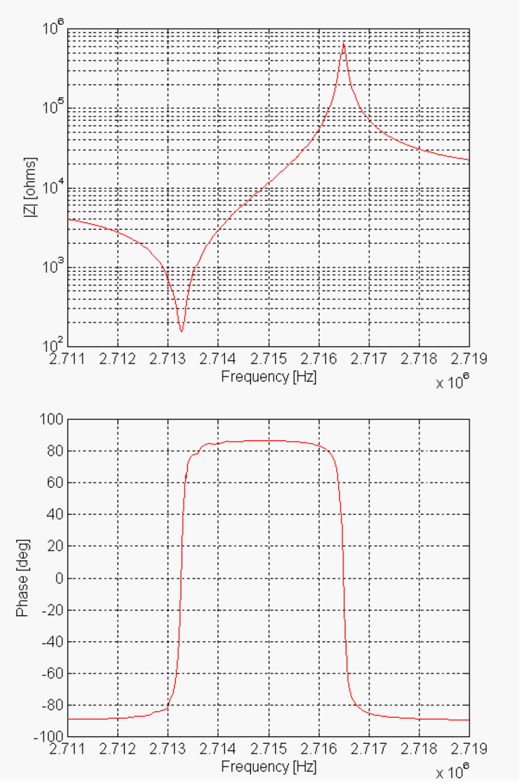

Fig 4: Quartz crystal impedance and phase

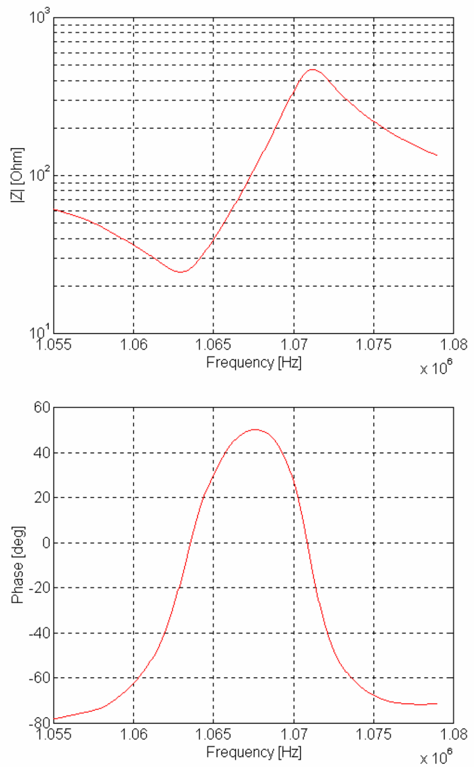

Fig 5: Piezoelectric Ceramic PZT impedance and phase

It can be seen that both the quartz and PZT disc well meet the required conditions to reach a stable oscillation at the given frequency. However, ceramic discs have a disadvantageous feature, namely that radial mode is much more dominant than the thickness mode, subsequently cuts must be made on them to suppress this mode.

We also analyzed some specimens of different periodicity and filling polymer 1-3 piezo-composites (fig. 6.) and compared the characteristics with the thick-film PZT transducers used in the design of the gas sensor. In this case the radial mode doesn't exist and they have relatively good impedance characteristics. Nevertheless, its use for the gas sensor cell design is limited to very high periodicity (near 100%), because at the given wave-mirror distances (cca. 166 μm) near-field emission characteristics of 1-3 piezocomposites take impossible stationary waves [9].

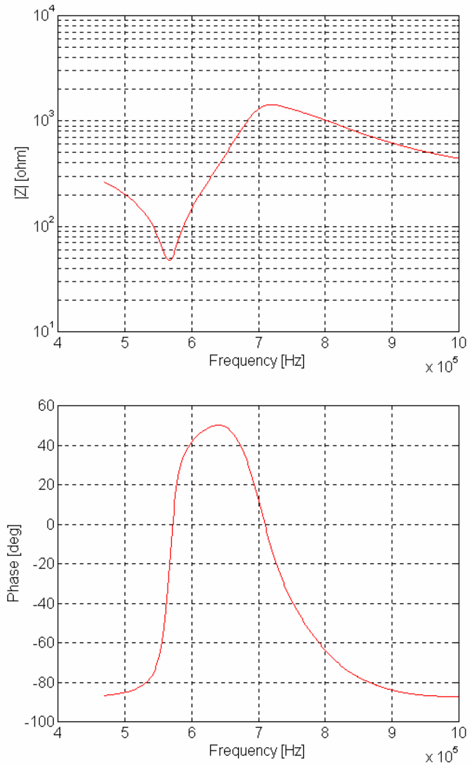

Fig. 6: 1-3 Piezocomposite impedance and phase

Thick film PZT ceramic fabrication and polarization was modified some times in order to improve their impedance characteristics. Figure 7 shows that in this case reactance features are much poor than in the other studied cases. From the point of view of the inductance presented by different piezoelectric devices, between series resonant frequency and the antiresonance, one can see that while quartz crystal it is around some tenths of millihenry, PZT discs are less than one millihenry and for 1-3 piezocomposites it is of some tenths of microhenry. Nevertheless in the case of the thick film devices it is in the order of some nanohenry. Additionally, the effective resistance of the transducer is very low.

Possibly it is due to the greater internal and superficial porosity of this material, as well as due to the not really good adhesion between painted layers. This fact greatly difficult the job of making a highly stable oscillator as it is needed. Presently, authors are improving these characteristics changing the used materials composition. Printing characteristics like wettability and adhesion have to be studied in more detail.

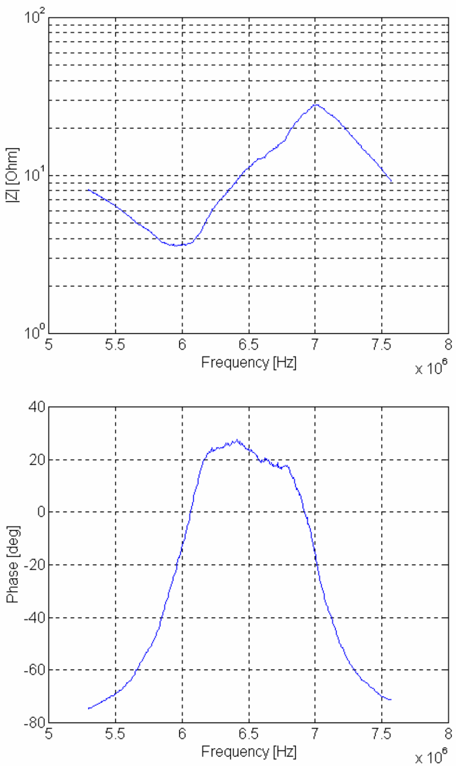

Fig. 7. Thick Film PZT impedance and phase

Conclusions

The analysis of the thick film piezoceramics developed leads us to the following conclusions:

1. The thick film obtained is piezoelectric active, but the reactance characteristics of the ceramics must be improved changing used materials composition and tightening.
2. Dimensions obtained for the thick film cell cavity are adequate to reach the resonance parameter **l** (about 0.166 mm in air).
3. Effects of the other resonances modes over the frequency stability of the sensor were evaluated in piezoceramics, piezo-composites and thick film piezoceramics.
4. A very stable electronic circuit must be used to compensate low **Q** and low inductance of thick-film piezoceramics.

Acknowledgments

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