CIRCUIT FOR CONTINUOUS MOTIONAL SERIES RESONANT FREQUENCY AND MOTIONAL RESISTANCE MONITORING OF QUARTZ CRYSTAL RESONATORS BY PARALLEL CAPACITANCE COMPENSATION.

PACS REFERENCE: 4338FX

Arnau Antonio; Sogorb Tomás; Jiménez Yolanda; Calvo Ernesto*; Otero Marcelo*
Laboratorio Integrado de Bioingeniería, Centro de investigación e innovación en Bioingeniería,
Universidad Politécnica de Valencia, Camino de Vera s/n.
Valencia 46022
España
Tel: 96 3877007 Ext:6041
Fax: 963879609
E-mail: aarnau@eln.upv.es
* INQUIMAE- Departamento de Química Inorgánica, Analítica y Química Física, Universidad de Buenos Aires, Pabellón 2, Ciudad Universitaria AR-1428 Buenos Aires, Argentina

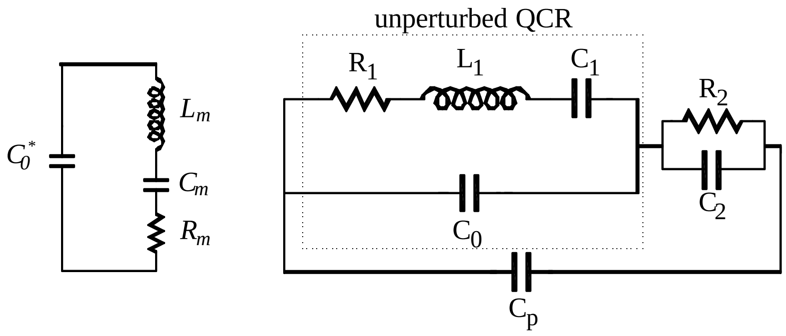
ABSTRACT
A deep analysis of the problem associated to interface circuits for quartz-crystal-microbalance sensors reveals that the so-called static capacitance of the sensor is one of the elements which make the use of oscillators more critical for sensors applications. A phase-locked-loop based circuit designed for compensating the parallel capacitance in quartz crystal resonator sensors is presented. This circuit permits the calibration of the external circuitry to the sensor and provides a continuous measurement of the motional series resonant frequency and motional resistance. An extension and automation of the proposed system for multiple sensor characterization is introduced. Experimental results are also shown.

INTRODUCTION

The quartz crystal microbalance (QCM) is commonly used as sensor both in gaseous and liquid media. The behavior of quartz crystal resonator (QCR) sensors in liquids differs essentially from that in gaseous media, mainly because of the increase in the damping and to the more important role which plays the parallel capacitance in the impedance of the sensor. Impedance Analyzers (IA) are typically used in laboratory for an electrical characterization of the sensor around resonance. These systems characterize the sensor in isolation and they permit the exclusion of parasitic influences by calibration, their disadvantages are the high cost and large dimensions. Special oscillators have been designed to provide a measurement related to the sensor damping [1]. These systems are commonly use principally due to the low expense of the circuiting and to the mayor adaptability for in situ measurements in comparison with impedance analyzers. The problems associated to oscillators are treated in this paper, then a circuit which retains the simplicity of oscillators while overcoming their limitations is presented. Finally, some experimental measurements done with that system are presented as well.

THEORY

The shear strain induced in a loaded AT cut quartz when an alternating-current (AC) voltage is applied across the crystal, generates a transversal acoustic wave propagating through the quartz to the contacting media. The mechanical interaction between the resonator and the contacting media influences the electrical response of the device and makes possible the use of the resonator as a sensor. The electrical impedance of quartz resonator in liquids around the resonant frequency relative to a given mode number can be approximated by an extended Butterworth-Van Dycke (EBVD) circuit model (Fig.1a). This circuit comprises a capacitance C_o^* in parallel with a series branch (R_m, L_m and C_m) that models the so-called motional impedance Z_m arising from electrically excited mechanical motion in the loaded quartz [2]. The capacitance C_o^* includes both the crystal capacitance and the parasitic capacitance external to the sensor. From this model, the most important parameters to characterize the sensor response are: the motional resistance R_m and the motional series resonant frequency (MSRF) $f_s = 1/(2\pi\sqrt{L_m C_m})$.



(a) (b)

Fig. 1.- (a) Equivalent circuit of a loaded quartz-crystal-resonator. (b) Circuit for simulating a quartz resonator sensor

PROBLEM ASSOCIATED TO OSCILLATORS FOR HEAVY LOADED QUARTZ-RESONATORS

A sensor-controlled oscillator tracks the frequency for a certain sensor impedance phase. When the resonator properties are altered due to loading effects, the phase of the resonator changes and the frequency shifts to produce a sensor phase that fits the oscillation condition. The MSRF only depends on L_m and C_m components of the motional branch, but the phase of the complete resonator also depends on the specific values of the motional resistance R_m and on the parallel capacitance C_o^*, in this way changes in these parameters produce erroneous changes in the oscillating frequency. These erroneous changes would not take place if the parallel capacitance was precisely compensated and the phase of the sensor for oscillating condition was 0º. Fig.2b represents the phase of the sensor admittance ($Y=j\omega C_o^*+1/Z_m$) versus the frequency for a 10MHz AT cut QCR loaded with different media. It is considered that the quartz is one-face in contact with the polymer YO2 with viscoelastic properties $G_1'=2.1\times10^5$ *Pa* and $G_1''=2.1\times10^6$ *Pa*. Several thicknesses of the first layer, h_1, were considered. The medium contacting the first layer is air. Fig.2a shows in the x-axis the relative frequency errors between oscillating frequency shifts, Δf^{φ},and MSRF shifts, Δf_{MSRF}, for each medium, and for different values of the admittance phase. These frequency shifts are relative to the MSRF in the unperturbed state, which can be considered independent on the sensor phase. These relative errors are mathematically expressed as follows:

$$ {}_{\Delta f}(\%) = \left| \frac{\Delta f^{j} - \Delta f_{MSRF}}{\Delta f_{MSRF}} \right| \times 100 = \left| \frac{f^{j}{}_{osc} - f_{MSRF}}{f_{MSRF} - f_{v}} \right| \times 100 $$

Where $f^{j}{}_{OSC}$ is the oscillating frequency determined by a certain phase of the sensor, f_v, is the MSRF of the sensor in the unperturbed state and f_{MSRF} is the MSRF of the loaded sensor. From this graph can be extracted that when an oscillator is used to monitor frequency shifts of quartz sensors only subject to inertial loads, the frequency shifts monitored are practically independent of the working phase of the sensor in the oscillator. However, when the quality factor changes during experiment the frequency shifts monitored are strongly dependent of the working phase of the sensor in the oscillator. This is illustrated in Fig.2a for two cases, working phases of 0º and -40º, corresponding to typical oscillator designs.

As can be observed, the sensor is able to provide the oscillating phase condition of -40º for all loads and the relative error is ranged between 10% and 25% depending on the load. However, it can be observed that for an oscillating phase condition of 0º, the relative frequency error increases while increasing the load damping, and a maximum error around 43% is reached for h_1 =0,8μm . A further increase in the damping of the load makes the sensor unable to provide an oscillating phase condition of 0º and the oscillation ceases.

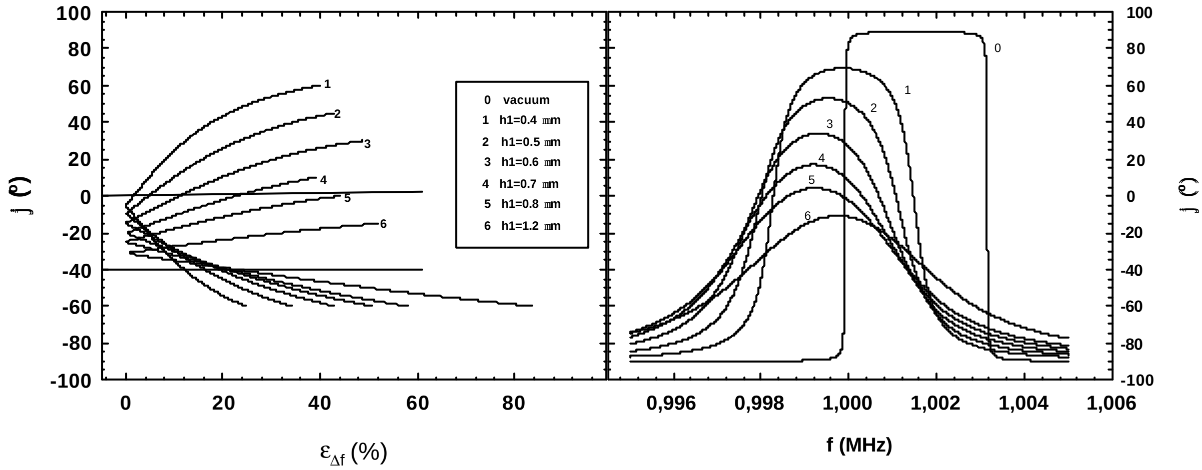


Fig. 2.- Diagram for illustrating the problem associated to oscillators as interface circuits for loaded QCRs. a) Relative frequency error between oscillating frequency shifts and MSRF shifts vs the phase of the sensor. b) Phase of the loaded QCR vs frequency.

After the described analysis, an electronic driver for quartz sensor applications should fulfill the following requirements:

- The electronic driver should operate like an oscillator providing analog signals closely related to the MSRF and motional resistance. However, an improvement in the accuracy in determining these parameters must be achieved in comparison to currently used oscillators.
- The sensor should be monitored in isolation. Then, the circuit must provide a mean for making a calibration of the external circuit to the sensor so that a continuous operation of the QCR at the MSRF can be assured.
- The parallel capacitance including possible stray capacitances in parallel must be eliminated or compensated by the system. Then, a mean for a precise determination of the parallel capacitance compensation must be provided by the system. It is also advisable that one of the quartz electrodes is grounded in order to minimize parasitic effects and to enable a proper sensor operation for electrochemical or biological applications.
- A possible extension of the circuit for multiple sensor characterization should be considered as well.

The authors have designed a new interface circuit for quartz sensor applications which fulfills the former requirements.

SYSTEM DESIGN

The proposed system shown in Fig.3 is based on a phase-locked-loop (PLL) configuration built around the zero-pass detectors *IC1*, *IC2*, the phase-frequency-detector (PFD), the low-pass filters (LPFs), the differential amplifier (DA1) and the active integrator filter (IF) connected to the input of a sinusoidal voltage-controlled crystal oscillator (VCXO) for closing the loop. This PLL circuit is calibrated for a lock-in condition that makes the phases of the signals at points A and B equal [3]. The parallel capacitance compensation is accomplished around the operational amplifier (OPA) A_1 which develops a voltage $u_A = [1+R_v/Z_m + j\omega C_r R_v]\, u_i$, where Z_m is the motional impedance associated to the motional branch and $C_r = C_o^*-C_v$ is the residual uncompensated parallel capacitance, which is zero when $C_v = C_o^*$. In this situation, the voltage signal u_A only depends on Z_m and will be in phase with the voltage signal u_B = u_i at the MSRF, which will be continuously tracked by the system. The cancellation of the capacitance C_o* can be accurately performed at a frequency double of the fundamental resonant frequency, where only static capacitive behavior of the sensor is expected. At this frequency the variable capacitor C_v is adjusted in such a way that the voltage u_j reaches the same value as in the lock-in condition. This indicates that the parallel capacitance C_o* has been compensated. At MSRF $Z_m = R_m$ and $u_A / u_B = 1 + R_v / R_m$. The circuit built around the logarithmic amplifiers *L1*, *L2* and the differential amplifier *DA2*, provides an output voltage $u_{Rm} = \alpha + \beta \log(u_A / u_B)$, where α and β are constants to be determined in a characterization procedure. Combining the former expressions, the voltage u_{Rm} permits a continuous motional resistance monitoring.

EXPERIMENTAL RESULTS

The system was calibrated and characterized by connecting ten different selected resistances, covering a range from 100 Ω to 5000 Ω, in place of the sensor. Values of α =-228.8mV and β=3420mV were obtained were obtained which minimized the sum of squared relative errors between the values of R_m obtained from the voltages u_{Rm} and the corresponding resistance values measured with an ohm-meter. In order to evaluate the reliability of the proposed system, we used the circuit shown in Fig.1b to simulate the loaded sensor. In this circuit, a series branch comprising a 10MHz unperturbed quartz resonator (modeled with C_o, R_1, L_1 and C_1) and a resistor R_2, is connected to a parallel capacitor C_p. The components used in this circuit were separately measured with an impedance analyzer carefully calibrated and their values were: C_o = 8.52 pF, R_1 = 6.86 Ω, L_1 =11.331 mH, C_1 = 22.3682 fF, C_p = 5.82 pF, R_2 = 100, 270, 510, 780, 1000, 1176, 1500, 2200, 3900 and 4700 Ω. A parasitic capacitance C_2 = 0.209 pF was measured as well. The proposed system was used to compensate the parallel capacitance C_p, to monitor the motional resistance R_1+R_2 and to monitor the phase-zero frequency of the series branch of the circuit, which should be close to the MSRF of the unperturbed QCR, $f_s = 1/(2\pi\sqrt{L_1 C_1})$.

An impedance analyzer was also used to extract the magnitude of the conductance peak of the simulated sensor ($1/(R_1+R_2)$) and the corresponding frequency (MSRF). Monitored frequencies from the proposed system and from the impedance analyzer are plotted in Fig.3 (upper panel) versus R_2. The equivalent motional resistances of the simulated sensor monitored with the proposed system and those provided by the impedance analyzer are shown in Fig.3 (lower panel) versus R_2. The expected resistances calculated as R_1 + R_2 have been also included for comparison. As can be observed the frequencies and motional resistance values monitored with the proposed system fit reasonably well to those obtained from the impedance analyzer. An error smaller than 3% was computed for all the resistances. A greater deviation from the expected resonant frequency of the unperturbed QCR is observed for higher resistance values. This deviation is not due to an uncompensated capacitance C_r, since the same tendency is observed in the maximum conductance frequency which does not depend on the parallel capacitance C_p. Therefore, this deviation is produced by the parasitic capacitance C_2, which will not appear in a real sensor.

Calibration and parallel capacitance compensation procedures can be automated for improving the system which can be easily extended to multi-sensor applications.

Preliminary Measurement

An AT-cut 10 MHz quartz crystal was used as QCM sensor. The quartz crystals were mounted by concentric o-ring seal into a flow-through cell to provide contact with only one side of the quartz crystal to the liquid. The flow-through cell was Teflon-made with owner design.

The QCM sensor was connected to the impedance analyzer HP4292A. The magnitude of the conductance and the susceptance were monitored and the resonance frequency was measured. A personal computer was used for the collection of the data via IEEE-488 interface. The data acquisition program was written in Visual Basic. Time between measurements was two seconds.

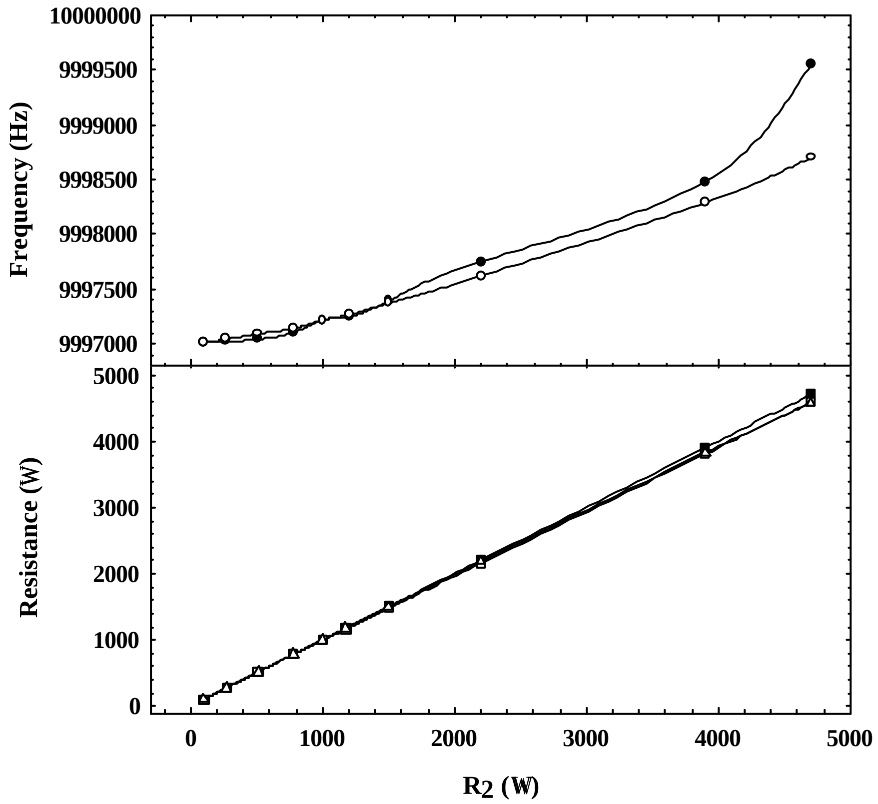


Fig. 3.- Experimental results obtained with the system and with the impedance analyzer
maximum conductance frequency monitored with the impedance analyzer.
frequency monitored with the proposed circuit.
$R_1 + R_2$
resistance monitored with the proposed system
Δ equivalent motional resistance monitored with the impedance analyzer.

The quartz crystal electrode surface was prepared for thiolation with cistamine during one hour, rinsing with buffer phosphate (PBS) 10 mM with pH 7.2-7.5 0.135 M of NaCl.

The flow-through cell was connected to pump system and was applied the conjugate OVA-CNA 500nM in PBS buffer during an hour until the resonance frequency of the QCM sensor was stabilized. It is shown in Fig.4. Rinsing again with PBS and was applied BSA for covering up any hole between conjugate. The BSA 0.1% solution in PBS was applied until the resonance frequency of QCM sensor was stabilized. Rinsing again with PBS and the QCM sensor was ready for experimental immunoassay.

The antibody LIB-CNH36 solution 200nM was supplied by a modified injection valve at specific flow rate. The resonance frequency as a time function was shown in Fig.5.

CONCLUSION

The practical experiments made for system evaluation show good agreement with IA measurements with IA measurements which makes this system ver useful for sensor application as it has been shown in preliminary experiments where the application of IA is cumbersome. At the same time the system design ha the great adaptability of typical oscillators while overcoming their inaccuracy in resistance and frequency determination.

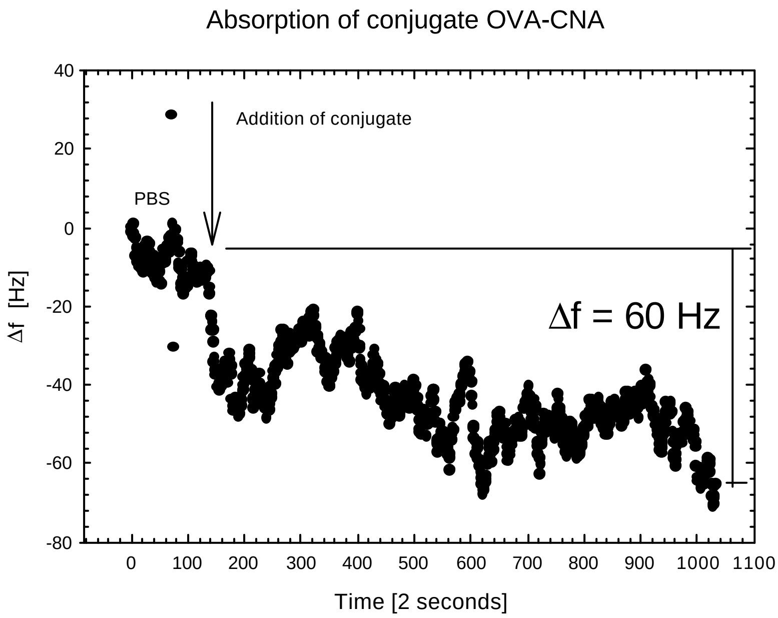


Fig. 4.- Absorption of conjugate OVA-CNA

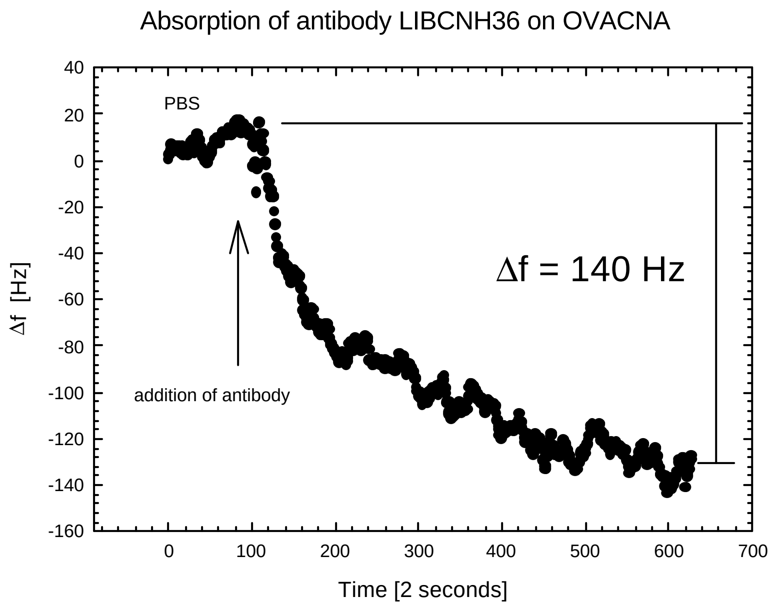


Fig. 5.- Absorption of antibody LIBCNH36 on OVACNA

REFERENCES

1 EICHELBAUM, F., BORNGRÄBER, R., SCHRÖDER, J., LUCKLUM, R., AND HAUPTMANN, P.: 'Interface circuits for quartz-crystal-microbalance sensors' *Rev. Sci. Instrum.*, 1999, **70**, (5) pp. 2537-2545

2 ARNAU, A., JIMÉNEZ, Y., AND SOGORB, T.: 'An Extended Butterworth-Van Dyke Model for Quartz Crystal Microbalance Applications in Viescoelastic Fluid Media', *IEEE Trans. Ultrason., Ferroelect., Freq. Contr.*, 2001, **48**, (5) pp. 1367-1382 (2001).

3 ARNAU, A., SOGORB, T., AND JIMÉNEZ, Y.: 'A continuous motional series resonant frequency monitoring circuit and a new method of determining Butterworth-Van Dyke parameters of a quartz crystal micobalance in fluid media', *Rev. Sci. Instrum*. 2000, **71**, (6) pp. 2563-2571.