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Application of the impedance transformation on transmission lines to electrochemical microbalance

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Abstract. — The purpose of this paper is to propose an analysis of stacked mechanical resonator designs in terms of microwave network approach. The application to electrochemical quartz crystal microbalance (EQCM) is achieved where we have privileged the fundamental impedance transformation equation on transmission line relating the impedance at any point of the line to that at an other point of the line. From this systematic procedure applied to a quartz crystal with simultaneous mass loading and liquid loading we have derived the impedance expression and the near-series-resonance characteristics of the loaded crystal resonator by means of a multilayer model. The effect of thickness of a mass layer and of a contacting Newtonian liquid on real and imaginary parts of impedance is predicted for several deposited materials. A lumped equivalent circuit is obtained from this formulation and the proposed results are compared with recently published experiments. The analysis presented in the familiar context of distributed circuit theory make the formulation and the physical interpretation of various scattering problems easier.

1. Introduction.

The propagation and scattering of waves have been extensively studied in the areas of acoustics, electromagnetics and elastodynamics. These problems have received much attention in the area of microwave theory and the microwave methods (equivalent circuits, scattering representations, discontinuities, multiple reflections, coupled modes, mode expansion and so on) are concepts of great interest in acoustic wave applications [1, 2] such as acoustic composite resonators, quartz crystal microbalances, etc.

Fundamentally the analogy is close enough with apparatus of the order of magnitude of wavelength. We use this close analogy that present waves and we propose an original calculation which allows the analysis of behaviour of multilayered structures such as EQCM (electrochemical quartz crystal microbalance).

Among the theories developed to analyse quartz crystal microbalances (QCM), the theory based upon Mason equivalent circuit has been applied to many situations [3]. Crane and Fischer [4] extended the transmission line analogy of Williams and Lamb [5] to the application

In the present paper, with some simple notational changes, the acoustic field equations may be written into the form of transmission lines equations. This presentation will simplify the task of transferring to acoustics the analytical methodology and techniques which have been applied in microwave transmission lines. The method easy to carry out is based on the concept of impedance and impedance transformation on lines. The condition determining the behaviour of the acoustic field at a boundary between two media is the same as the behaviour of the tension $V$ and the current $I$ at a discontinuity in a transmission line and all discussions of reflection coefficients, of composite lines, of impedance matching can be carried out. In each medium we have a direct and a reflected wave, a complex reflection coefficient and an electrical impedance $Z$, the ratio of $V$ to $I$. This impedance varies from point to point and it is continuous at a boundary between two media. It is easy to calculate the impedance at any point in the system. We have take the advantage of the study of propagation and of impedance to get a solution of the problem of multiple reflections in layered media. In particular, this methodology facilitates the calculation of the electrical input impedance for arbitrary acoustic load in EQCM and provides in sight into the role of key parameters, mass loading and liquid loading, which affect the EQCM response. On the other hand, this mathematical model is not limited to thin deposits in EQCM and it is possible to solve many kinds of acoustic problems with distributed electrical circuit model.

2. Electromagnetic waves and transmission line analogy.

In electromagnetic wave propagation, the electric field $E$ and the magnetic field $H$ are regarded as fundamental and are chosen as the basic variables. In addition, two auxiliary field vectors are introduced, namely, the electric displacement $D$ and the magnetic induction $B$ which are related to $E$ and $H$ through the characteristics of propagation material media by means of constitutive relations.

A variety of structures for the transport of electromagnetic energy in the form of a propagating wave has been developed such as coaxial line where the electric field $E$ is derived from the gradient of a scalar potential $V$. A close analogy exists between the propagation of plane waves in a homogeneous medium and the propagation of voltage $V$ and current $I$ along a transmission TEM (Transverse Electro-Magnetic) line such as a coaxial line.

In the acoustic problems the analogy between the forces $F$ (or the stress $T$) and particle velocities $v$ in an elastic medium with the voltages $V$ and currents $I$ on a line may be made also for propagating plane waves.

Among the quantities in acoustic equations, four other quantities may also be chosen by analogy to $E$, $H$, $D$, $B$, namely, the stress $T$, the particle momentum field $p$, the velocity $v$ and the strain $s$.

2.1 Electrical transmission line equations [12]. — The characterization of electrical transmission lines requires the evaluation of two fundamental line parameters, the characteristic impedance $Z_c$ of the line and the propagation constant $\gamma = \alpha + j\beta$ (attenuation $\alpha$, phase constant $\beta$) in terms of physical parameters and properties of dielectric and conductor materials used.

The equivalent circuit of a line of length $l$ and waves travelling in the $z$ direction is shown figure 1, where $dz$ is an infinitesimal line length.
Fig. 1. — Equivalent circuit of a line.

The lumped impedance \( Z_1 \) per unit line length and the lumped admittance \( Y_2 \) per unit line length are given by:

\[
Z_1 = \gamma Z_c \quad (1)
\]

\[
Y_2 = \frac{\gamma}{Z_c} \quad (2)
\]

Usually the differential equations describing the steady-state of wave on transmission line are Helmholtz equations whose solutions for voltage \( V \) and current \( I \) are:

\[
V = (A e^{-\gamma z} + B e^{+\gamma z}) e^{jwt} \quad (3)
\]

\[
I = \frac{1}{Z_c} (A e^{-\gamma z} - B e^{+\gamma z}) e^{jwt} \quad (4)
\]

where the fundamental line parameters are expressed by:

\[
\gamma = (Z_1 Y_2)^{1/2} \quad (5)
\]

\[
Z_c = \left( \frac{Z_1}{Y_2} \right)^{1/2} \quad (6)
\]

2.2 Generalized Parameter Sets of Line. — The behaviour of a line can also be characterized in terms of an impedance matrix \([Z]\), an admittance matrix \([Y]\), an hybrid matrix \([h]\), a chain matrix \([c]\) or a scattering matrix \([s]\), together with boundary conditions at the line output \((V_2, I_2)\) and at the line input \((V_1, I_1)\) for a line length \(\ell\). However, one parameter set may be more convenient than others in a particular analysis and design problem.

Among these representations, the chain matrix is very suitable for the analysis of cascaded circuits or cascaded lines and allows to write the propagation equations in hyperbolic function form as \([13]\):

\[
\begin{bmatrix}
V_1 \\
I_1
\end{bmatrix} =
\begin{bmatrix}
\cosh \gamma \ell & Z_c \sinh \gamma \ell \\
\frac{1}{Z_c} \sinh \gamma \ell & \cosh \gamma \ell
\end{bmatrix}
\begin{bmatrix}
V_2 \\
I_2
\end{bmatrix} \quad (7)
\]

where it is usual to take the location of the load impedance \(Z_s\) of the line as the reference point and to consider the sending end as being to the left of this reference point.

2.3 Impedance Transformation on Lines. — The general expression of the impedance \(Z_d\) at the distance \(d\) of the load \(Z_s\) may be obtained from equation \((7)\). For lines with losses and
without losses, $Z_d$ is given respectively by:

$$Z_d = Z_\circ \frac{Z_s + Z_t \tanh \gamma d}{Z_s + Z_t \tanh \gamma d}$$  \hspace{1cm} (8)$$

$$Z_d = R_\circ \frac{Z_s + jR_e \tan \beta d}{R_\circ + jZ_s \tan \beta d}$$  \hspace{1cm} (9)$$

where $\gamma = j\beta$ and $Z_\circ = R_\circ$ for lossless lines.

The impedance description, just as the scattering description of a propagation mode constitutes the complete description of the mode fields everywhere in terms of the incident and reflected amplitude at a single point.

In particular, the impedance formulation may be used to obtain a rigorous generalized transverse resonance relation for the determination of electrochemical quartz microbalance properties.


One of the most important and simplest boundary value problems in acoustic is the case of a uniform plane wave perpendicularly incident upon a plane discontinuity surface as in many acoustic resonators. For arbitrary incidence angle and other problems of a more complex nature, the common features between electromagnetism and acoustics are still helpful in providing or in suggesting approximation procedures [14].

3.1 Acoustic line parameters. — First, the transmission line analogy is applied to an AT cut quartz because the AT cut is the used cut in the EQCM. It is assumed that the quartz plate is an infinite plane plate. Also in practice, the lateral dimensions are usually large compared with the acoustic wavelength. The wave is travelling in the thickness direction which is taken as z direction. A single mode of vibration without coupling to other modes is assumed. For only one of the displacement eigendirections, a chain of transmission line may be used since the differential equations describing the steady-state shear waves are the Helmholtz equations.

Equations (3) and (4) are similar to the harmonic solutions of the acoustic Helmholtz equations:

$$- T = (A^- e^{-\gamma m z} + B^+ e^{+\gamma m z}) e^{j\omega t} \hspace{1cm} (10)$$

$$v = \frac{1}{Z_m} (A^- e^{-\gamma m z} - B^+ e^{+\gamma m z}) e^{j\omega t} \hspace{1cm} (11)$$

In our work, negative stress $T$, particle velocity $v$ are respectively identified with $V$, $I$ of electrical transmission line equations in the direct analogy of impedance type. For an acoustic wave propagating in a material $m$, without losses, the distributed elements of the equivalent line are $Z_1 = j\rho_m \omega$ and $Y_2 = j\omega c_m^{-1}$, and acoustic line parameters $\gamma_m$, $Z_m$ are given by:

$$\gamma_m = (Z_1 Y_2)^{1/2} = j\omega \left( \frac{\rho_m}{c_m} \right)^{1/2} = j \frac{\omega}{v_m} \hspace{1cm} (12)$$

$$Z_m = \left( \frac{Z_1}{Y_2} \right)^{1/2} = (\rho_m c_m)^{1/2} = \rho_v v_m \hspace{1cm} (13)$$

where $v_m = (c_m/\rho_m)^{1/2}$ is the acoustic wave velocity, $\rho_m$ the mass per unit volume, $c_m$ the stiffness coefficient for the considered shear mode in the material $m$ and $Z_m$ the equivalent characteristic impedance of the acoustic line. Because of the non-physical
nature of \( V \) and \( I \), the equivalent transmission line impedance \( Z_m \) in these equations may therefore be chosen in different ways to suit particular problems. In our analogy, \( Z_m \) is found by considering the ratio of \(-\frac{T}{p}\) for an infinite fictitious line without reflection. In that case this characteristic impedance \( Z_m \) is an acoustic impedance per unit cross area and depends only on material constants \( \rho_m \), \( c_m \) associated with the considered acoustic line.

For the sake of generality the quantity \( c_m \) can be defined as an effective complex shear modulus: 
\[
\tilde{c}_m = c_m + \frac{\varepsilon^2_m}{\varepsilon_m} + j \omega \eta_m
\]
where \( c_m + \frac{\varepsilon^2_m}{\varepsilon_m} \) is the stiffened shear modulus for a piezoelectric media and \( j \omega \eta_m \) points the losses (the quantities \( \varepsilon_m \), \( \varepsilon_m \), \( \eta_m \) are the piezoelectric constant, the dielectric constant, and the viscosity of medium \( m \) respectively).

3.2 DISTRIBUTED ELEMENT CIRCUIT OF A MULTILAYER STRUCTURE: SINGLE MODE REPRESENTATION OF A LOADED QUARTZ. — The previous transmission line results may be applied in order to examine the behaviour of a lossless medium, the quartz \( (m = q) \), characterized by \( \rho_q \), \( \nu_q \), \( h_q \) and a lossless coating \( (m = c) \) characterized by \( \rho_c \), \( \nu_c \), \( h_c \), as in figure 2.

![Fig. 2. — A multilayer structure.](image)

The model behaves two cascaded transmission lines terminated by short-circuits corresponding to stress-free boundary conditions. So the quartz is terminated at the interface quartz-coating in the plane CD by a load impedance \( Z_{CD} = j \rho_c \nu_c \tan \frac{\omega h_c}{\nu_c} \) obtained from (9).

The resonance condition is also given from (9) by writing that \( R_c = \rho_q \nu_q \) and that the impedance \( Z_{AB} \) must be equal to zero at the distance \( h_q \) of the interface-quartz coating. Thus, the mechanical (parallel) resonance condition is easily obtained:

\[
\rho_c \nu_c \tan \frac{\omega h_c}{\nu_c} + \rho_q \nu_q \tan \frac{\omega h_q}{\nu_q} = 0.
\]

This result corresponds to the well established equation of the layered resonator [7, 20, 21].

An idealized multilayer quartz crystal is shown in figure 3a. Two transmission lines \( E_1 \), \( E_2 \) (line parameters \( Z_{e1}, \gamma_{e1}, Z_{e2}, \gamma_{e2} \)) describe the electrodes of the resonator. \( Z_q \) and \( \gamma_q \) are the parameters of the equivalent line of quartz and \( h_q \) is the quartz thickness. Lines \( E_1 \) and \( E_2 \) are loaded by medium \( m \), usually air or vacuum (liquid in electrochemical quartz crystal microbalance). If the loading medium \( m \) has high losses and thickness \( h_m \gg \lambda_m \), \( \lambda_m \) being the acoustic wavelength in the medium \( m \), the medium can be considered as semi-infinite since there is no reflected wave in that medium. Then lines \( E_1 \) and \( E_2 \) are loaded by \( Z_m \) the acoustic characteristic impedance of the medium \( m \) (Fig. 3b).
The two acoustic ports are coupled with the electrical port \((V_3, I_3)\) by means of an electrical circuit [10] connected to the center of the quartz transmission line. This electrical circuit offers advantage in applications where several circuits are cascaded acoustically. The role of the electrical and mechanical parts of the circuits are well distinguished and the new circuit facilitates the calculation of electrical input impedance for arbitrary acoustic loads in EQCM.

In this circuit \(C_0 = \varepsilon_q \frac{A}{\varepsilon_{q} Z_q}\) is the static capacitance (\(\varepsilon_q\) is the quartz permittivity, \(A\) the electrode area of the crystal). The piezoelectricity is modeled by a piezoelectric transformer of turn ratio \(N\) and by the element \(X\). In transducer theory it is customary to use total force \(F\) (rather than stress \(T\)) as the « acoustic voltage » variable at the disk terminals. With the stress as « acoustic voltage » variable, \(N^2\) and \(X\) are expressed as a function of \(Z_q = \rho_q v_q\), as:

\[
N^2 = A \left( \frac{\varepsilon_q \omega Z_q}{2 \varepsilon_q \sin \frac{\omega h_q}{2 v_q}} \right)^2
\]  
\[X = \frac{\varepsilon_q^2}{A \varepsilon_q^2} \frac{\omega h_q}{\omega^2 Z_q} \sin \frac{\omega h_q}{v_q}
\] 

Fig. 3. — a) Multilayer quartz crystal. b) Electrical model of a quartz loaded by semi-infinite media. The transformer of ratio \(1 : N\) characterizes the indirect piezoelectric effect.
4. Application to electrochemical quartz crystal microbalance (EQCM).

The composite resonator associated to an EQCM consists of three layers: the quartz crystals, the deposited mass layer, and a contacting liquid. A single thickness shear mode is considered.

The electrodes are highly conducting and have negligible mass compared to the deposited mass (the acoustic effect due to the electrode thickness will be assumed to be negligible). One of the quartz surface is air phase and is not affected by external forces (Fig. 4).

\[ \begin{align*}
\rho_q & \quad v_q \\
\rho_f & \quad v_f \\
\rho_l & \quad v_l
\end{align*} \]

Fig. 4. — A quartz loaded by a mass layer and a contacting liquid.

In the general case, the three layers have losses and the continuity of shear stress and particle displacement at the boundaries between the different layers of the EQCM may be solved from the impedance transformation equation (8).

4.1 EQCM IMPEDANCE. — The EQCM equivalent circuit containing three cascaded acoustic lines is shown in figure 5. \( Z_r, \gamma_f \) are the line parameters of the mass layer (film of thickness \( h_f \)), in contact with one electrode and \( Z_f, \gamma_f \) the line parameters of semi-infinite liquid medium.

\[ \begin{align*}
Z_q & \quad \gamma_q \\
Z_f & \quad \gamma_f \\
Z_l & \quad \gamma_l
\end{align*} \]

Fig. 5. — Equivalent circuit for an EQCM.
Short-circuit boundary condition is applied at the other quartz electrode which is assumed to be mechanically free (vacuum or atmosphere).

The impedance $Z_{EF}$ at port EF is obtained from (8) with $Z_s = Z_{GH} = Z_t$ for a semi-infinite liquid medium.

$$Z_{EF} = Z_t \frac{Z_t + Z_t \tanh \gamma_t h_f}{Z_t + Z_t \tanh \gamma_t h_f}$$  \hspace{1cm} (17)

At port CD, the impedance $Z_{CD}$ is given from [8] and [18]

$$\frac{1}{Z_{CD}} = \frac{1}{Z_q} \left[ \coth \gamma_q \frac{h_q}{2} + \frac{Z_q + Z_{EF} \tanh \gamma_q h_q \gamma_q h_q}{Z_{EF} + Z_q \tanh \gamma_q h_q} \right]$$ \hspace{1cm} (18)

$$Z_{CD} = \frac{Z_q}{2} \left[ \frac{Z_q + Z_{EF} \tanh \gamma_q h_q}{Z_q + Z_{EF} \coth \gamma_q h_q} \right]$$  \hspace{1cm} (19)

The electrical impedance $Z_{EQCM}$ of quartz crystal microbalance at port AB is expressed as:

$$Z_{EQCM} = \frac{1}{j\omega C_0} + jX + \frac{Z_q}{2N^2} \left[ \frac{Z_q + Z_{EF} \tanh \gamma_q h_q \gamma_q h_q}{Z_q + Z_{EF} \coth \gamma_q h_q} \right]$$ \hspace{1cm} (20)

For a lossless quartz, $Z_{CD}$ and $Z_{EQCM}$ are reduced to:

$$Z_{CD} = \frac{Z_q}{2} \frac{Z_{EF} + jZ_q \tan x}{Z_q - jZ_{EF} \cot g 2x}$$ \hspace{1cm} (21)

$$Z_{EQCM} = \frac{1}{j\omega C_0} \left[ 1 - K^2 \frac{\tan x}{x} B \right]$$ \hspace{1cm} (22)

where $x = \frac{\omega h_q}{2 \nu_q}$ and $K^2 = \frac{\varepsilon^2_q}{\varepsilon_q c_q}$ is the electromechanical coupling constant of the quartz crystal for the considered vibration mode. $B$ is given by the following equation:

$$B = \frac{1 - j}{2 \frac{Z_{EF}}{Z_q} \tan x} \frac{Z_{EF}}{1 - j \frac{Z_{EF}}{Z_q} \tan 2x}$$ \hspace{1cm} (23)

In the absence of the mass layer and liquid layer, $Z_{EQCM}$ is reduced to the well-known expression [15] of the impedance $Z_Q$ of the infinite lossless quartz plane plate in vacuum, with stress-free boundary conditions at the crystal faces:

$$Z_Q = \frac{1}{j\omega C_0} \left[ 1 - K^2 \frac{\tan x}{x} \right]$$ \hspace{1cm} (24)

The expression of impedance $Z_{EQCM}$ allows a quantitative analysis of the EQCM and a
comparison with the curves obtained from an impedance analyzer. This analyzer gives modulus and argument of impedance or admittance versus frequency. The resonance conditions are deduced from these different curves.

Simultaneous effects of mass loading and liquid loading may be analysed from $Z_{\text{EQCM}}$ and in particular from motional impedance $Z_M$ of loaded quartz crystal, deduced from $Z_{\text{EQCM}}$ (in order to avoid any confusion with the characteristic impedance $Z_m$ of the material $m$, the motional impedance of the crystal is noted $Z_M$).

4.2 MOTIONAL IMPEDANCE $Z_M$ OF LOADED QUARTZ CRYSTAL. — The impedance expression $Z_{\text{EQCM}}$ may be converted into lumped-elements of an equivalent circuit whose circuit elements are related to physical properties of the mass layer, of the liquid and of the quartz.

The elements of a motional arm connected in parallel with the capacitor $C_0$, representative of the static capacitance between the crystal electrodes, may be obtained from the motional impedance $Z_M$ which is related to $C_0$ and $Z_{\text{EQCM}}$ by the relation

$$\frac{1}{Z_M} = \frac{1}{Z_{\text{EQCM}}} - j\omega C_0$$

$$Z_M = \frac{j}{\omega C_0} \left[ 1 - \frac{1}{K^2 \frac{\tan x}{x} B} \right].$$

For a quartz resonator driven at the series-resonance frequency corresponding to $x$ value close to $\frac{\pi}{2}$, $B$ is reduced to:

$$B = \left[ 1 - j \frac{Z_{\text{EF}}}{Z_q \tan 2x} \right]^{-1}$$

$$Z_M \approx \frac{j}{\omega C_0} \left[ 1 - \frac{1 - j \frac{Z_{\text{EF}}}{Z_q \tan 2x}}{K^2 \frac{\tan x}{x}} \right]$$

$$Z_M \approx Z_{M_0} - \frac{Z_{\text{EF}} x}{Z_q \omega C_0 K^2 \tan x \tan 2x}$$

where $Z_{M_0}$ is the motional impedance of the quartz crystal in vacuum and where the additional term is $Z_{M_a}$, the motional additional impedance which includes the effect of the liquid loading and mass loading.

Hence in the resonance vicinity, the additional impedance is reduced to:

$$Z_{M_a} \approx \frac{h_q^2}{4 v_q C_0 K^2 Z_q} = \frac{h_q^2}{4 A e_q^2} Z_{\text{EF}}.$$  

4.3 EFFECTS OF SIMULTANEOUS MASS LOADING AND NEWTONIAN LIQUID LOADING. — In Newtonian liquids, the line parameters are:

$$\left( Z_t \right)_N = (j\omega \rho_t \eta_t)^{1/2} = (1 + j \frac{\eta_t}{\delta_t})$$

$$\left( \gamma_t \right)_N = \left( j\omega \frac{\rho_t}{\eta_t} \right)^{1/2} = [1 + j \frac{\eta}{\delta_t}]$$
where $\delta_t$ is the viscous penetration depth:

$$\delta_t = \left(\frac{2 \eta t}{\rho t \omega}\right)^{1/2}$$  \hspace{1cm} (33)

The motional additional impedance $Z_{Ma}$ is given from (30) and (17) by:

$$Z_{Ma} = Z_f \frac{h^2_q}{4 A e^2_q} \frac{(1 + j) \eta t}{\delta_t} + Z_f \tanh \gamma t h_t$$  \hspace{1cm} (34)

Assuming a lossless film, $Z_{Ma}$ may now be written as:

$$Z_{Ma} = \frac{h^2_q \rho t \nu f}{4 A e^2_q} \frac{(1 + j) \eta t + j \rho t \nu f \tan \omega h_t}{\rho t \nu f + j (1 + j) \eta t \delta_t \tanh \omega h_t}$$  \hspace{1cm} (35)

and for $\frac{\eta t}{\delta_t \rho t \nu f} \tan \omega h_t \ll 1$, the motional additional impedance $Z_{Ma} = R_{Ma} + jX_{Ma}$ shows a motional resistance $R_{Ma}$ and a motional reactance $X_{Ma}$ given by:

$$R_{Ma} \approx \frac{h^2_q}{4 A e^2_q} \frac{\eta t}{\delta_t} \left(1 + \tan^2 \frac{\omega h_t}{\nu f} + \frac{\eta t}{\delta_t \rho t \nu f} \tan \frac{\omega h_t}{\nu f}\right)$$  \hspace{1cm} (36)

$$X_{Ma} \approx \frac{h^2_q}{4 A e^2_q} \left(\frac{\eta t}{\delta_t} + \rho t \nu f \tan \frac{\omega h_t}{\nu f} - \left(\frac{\eta t}{\delta_t}\right)^2 \frac{1}{\rho t \nu f} \tan \frac{\omega h_t}{\nu f}\right).$$  \hspace{1cm} (37)

For a thin film $\tan \frac{\omega h_t}{\nu f} = \frac{\omega h_t}{\nu f}$, the previous equation exhibits clearly the additional motional inductances $L_{Mf}$ due to Newtonian liquid loading and $L_{Mi}$ due to mass loading:

$$L_{Mf} = \frac{h^2_q}{4 A e^2_q} \frac{\eta t}{\delta_t} \omega = \frac{h^2_q}{4 A e^2_q} \left(\frac{\rho t \eta t}{2 \omega}\right)^{1/2}$$  \hspace{1cm} (38)

$$L_{Mi} = \frac{h^2_q}{4 A e^2_q} \rho t h_t.$$  \hspace{1cm} (39)

These expressions are in good agreement with the model of Nakamoto et al. [3] and with the model of Martin et al. [19]. They lead to the equivalent lumped circuit of the quartz (Fig. 6) simultaneous loaded by liquid and mass where $L_1$, $C_1$, $R_1$ are the motional elements of quartz in vacuum and where the additional motional resistance $R_{Ma}$ is given from the equation (36):

$$R_{Ma} \approx \frac{h^2_q}{4 A e^2_q} \frac{\eta t}{\delta_t} = \frac{h^2_q}{4 A e^2_q} \left(\frac{\rho t \eta t \omega}{2}\right)^{1/2}$$  \hspace{1cm} (40)

If $L_{Mf} + L_{Mf} \ll L_1 = \frac{h^3 q \rho q}{8 A e^2_q}$, the fractional change of the series-resonance frequency $f_s$, due to
the mass loading and to the liquid loading is:

\[
\frac{\Delta f_s}{f_s} \approx - \frac{L_{M1} + L_{Mf}}{2 L_1}, \\
\frac{\Delta f_s}{f_s} \approx - \frac{1}{h_q \rho_q} \left[ \rho_1 h_1 + \left( \frac{\eta f \rho \ell}{4 \pi f_s} \right)^{1/2} \right].
\]

The first term, in good agreement with the Sauerbrey results [6], is the contribution of the low mass loading. The second term is the contribution of the liquid loading. It is in good agreement with the result of Kanazawa et al. [17] and with the results of Reed et al. [21].

For thick films the crossed terms of equations (36) and (37) must be taken into account. So for a more general analysis, \( R_{Ma} \) and \( X_{Ma} \) must be deduced from equation (35) and the frequency \( f_r \) at which \( Z_{EQCM} \) is a pure resistance having a low value, is:

\[
f_r = f_s \sqrt{\frac{1}{1 + \frac{L_1}{L_{Ma}}}} \left[ \frac{1}{1 + \frac{C_0 (R_1 + R_{Ma})^2}{2 (L_1 + L_{Ma})}} \right]
\]

where \( L_{Ma} \) is the additional inductance deduced from \( X_{Ma} \) (37) and where \( f_s \) is the series resonance frequency usually defined as \( \frac{1}{2 \pi (L_1 C_1)^{1/2}} \), neglecting energy dissipation in the quartz crystal.

5. Application to electrochemical deposits in an aqueous medium: numerical results and discussion.

The quartz crystal is an AT cut plane parallel plate vibrating (Fig. 7) at a resonance frequency close to 6 MHz and with the following characteristics: \( h_q = 0.28 \) mm, plate diameter.
\( \varnothing = 16 \) mm, electrode diameter \( \varnothing_e = 5 \) mm, \( e_q = 9.65 \times 10^{-2} \) C/m\(^2\), \( L_1 = 50 \) mH, 
\( C_1 = 15 \) fF. The crystal oscillates in a pure shear mode, the direction of motion of crystal being exactly coplanar with the faces of the crystal disk. The quartz has one face in contact with air phase and the other face in aqueous medium. Since the used liquid is electrically contacting, the effective area of the electrode is enlarged [16].

Using «Mathematica» software, three kinds of deposits in water are simulated: gold, nickel and aluminium oxide having respectively a high, intermediate and low density. Their characteristics are given in table I and \( \rho_f = 998 \) kg m\(^{-3}\), \( \eta_f = 1 \times 10^{-3} \) Pa.s at 20 °C, \( \delta_f = 0.23 \) \( \mu \)m at 6 MHz.

Table I.

<table>
<thead>
<tr>
<th>Film deposit in aqueous medium</th>
<th>Aluminium oxide</th>
<th>Nickel</th>
<th>Gold</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho_f (10^3 ) kg m(^{-3}))</td>
<td>3.97</td>
<td>8.91</td>
<td>19.3</td>
</tr>
<tr>
<td>( Z_f (10^6 ) kg m(^{-2}) s(^{-1}))</td>
<td>24.6</td>
<td>26.7</td>
<td>23.2</td>
</tr>
<tr>
<td>( v_f (\text{m s}^{-1}) )</td>
<td>6 196</td>
<td>2 996</td>
<td>1 202</td>
</tr>
<tr>
<td>Wavelength (( \mu )m)</td>
<td>1 032</td>
<td>500</td>
<td>200</td>
</tr>
<tr>
<td>Thickness limit (( \mu )m)</td>
<td>50</td>
<td>25</td>
<td>10</td>
</tr>
</tbody>
</table>

The approximations worked-up in equation (27) and in equation (30) have been verified with this software for considered medium thickness. The effects of deposit thickness \( h_f \) in water on the real and imaginary parts of the \( Z_{Ma} \) impedance are evaluated at the series-resonance frequency 6 MHz with the equation (35) and with the simplified equations (38-40) for aluminium oxide deposit (Fig. 8), for nickel deposit (Fig. 9) and for gold deposit (Fig. 10).

![Fig. 8. — Effect of aluminium deposit thickness on \( Z_{Ma} \). (- - - -) From equation (35). (-----) From simplified equations (38-40).](image-url)
Fig. 9. — Effect of nickel deposit thickness on $Z_{M_3}$. (- - - -) From equation (35). (——) From simplified equations (38-40).

Fig. 10. — Effect of gold deposit thickness on $Z_{M_3}$. (- - - -) From equation (35). (——) From simplified equations (38-40).

Thus, from these curves it appears that, for thickness less than 10 $\mu$m, 25 $\mu$m, 50 $\mu$m (typically $h_f \leq \frac{\lambda_f}{20}$) for gold, nickel, aluminium oxide respectively, the simplified lumped circuit of figure 6 and equations (38-40, 42) are sufficient to predict the behaviour of an EQCM, accepting a 10 % relative error on real part of $Z_{M_3}$. Beyond equation (35) must be used. This last equation furnishes exact results whatever the film deposit thickness may be.

From previous equations, results are obtained with one face of the crystal in contact with water. This results show that the water viscosity adds an inductance $L_{Mt} = 15 \mu$H and a resistance $R_{Ma} \approx 577 \Omega$ to the mass loading inductance $L_{Mt}$ in the motional arm of the quartz circuit, for deposit thickness less than 1 $\mu$m for the three films. On the other hand $R_{Ma}$ increases as $(\rho_f \eta_f \omega)^{1/2}$ and reaches 706 $\Omega$ for a crystal in contact with a classical Watt bath for nickel deposit.

For deposit thickness $h_f = 10 \mu$m the additional motional resistance $R_{Ma}$ reaches the values 579 $\Omega$, 586 $\Omega$, 638 $\Omega$ for aluminium oxide, nickel and gold films respectively. These values reach 613 $\Omega$, 752 $\Omega$ and 5 970 $\Omega$ at $h_f = 40 \mu$m. For $h_f = \frac{\lambda_f}{4}$, $R_{Ma}$ and losses become high. The deposit in water act as antireflecting coating.

Hence for thick film the effect of $R_{Ma}$ must be taken into account in the evaluation of the resonant frequency and of the quality factor. $R_{Ma}$ must be evaluated from the real part of equation (34), i.e. from the real part of $\frac{h_f^3}{4 A e_q^2} Z_{EF}$. This analytical equation allows to evaluate
quality factor $Q$ and the frequency shift due to all ranges of deposit thickness in a liquid environment.

These theoretical results agree with experimental values obtained in passive measurements from a network analyser for liquid loading [18] and with results obtained with the matrix description [11].

On the other hand, these theoretical evaluations are also very interesting when the previous quartz crystal is used in an active device, i.e. in an oscillator circuit. Then it is possible to predict and to optimize the oscillation conditions of quartz crystal, with the ability of the crystal to continue to oscillate in contact with a viscous loading and a mass loading.

Thus the impedance transformation equation, usual in microwave, is well suited to analyse and to optimize the sensitivity of devices such as EQCM.

6. Conclusions.

Researchers in acoustics area can benefit from the existing literature in microwave for the solution of certain types of boundary value and scattering problems that have common mathematical models. The generalized approach based upon the microwave technique of impedance transformation in lines allows a general one-dimensional treatment of the layered piezoelectric resonator. In particular it is well suited for analysing the many different situations encountered in EQCM.

The introduction of acoustic voltage and current variables associated with acoustic wave has formal significance only. These two variables are related by a simple constant corresponding to the classical wave impedance of the propagation waveguide mode in the present paper. This specific acoustic impedance has been chosen as the characteristic impedance of line and could be used as normalization impedance in the expression of impedance transformation.

The electrical impedance $Z_{EQCM}$ is obtained by using impedance transformation of cascaded lines (quartz and mass layer) loaded by a liquid half-space (Newtonian liquid). The motional additional $Z_{Ma}$ of the quartz under loading conditions is derived from $Z_{EQCM}$ and provides a way to calculate resonant frequency shift and damping of the resonance as function of mass loading, liquid loading and thickness layers. This way is valid over all ranges of thickness. For thin films $h_l < \frac{\lambda_f}{20}$ the obtained expressions are identical with those established by preceding workers using other ways. For thick films the effect of crossed terms in motional resistance $R_{Ma}$ and in motional reactance $X_{Ma}$ is taken into account.

With this approach it is easy to solve many QCM problems only by changing the load $Z_{EF}$ connected to the acoustic port of the quartz resonator, and $Z_{EF}$ appearing in the expression of motional additional impedance is evaluated simply from the impedance transformation on a line of thickness $h_l$ loaded by a liquid half-space. The calculation of frequency shift $\Delta f$ and quality factor can be exactly achieved. The obtained equations furnish the shift of frequency according to changes in the essential parameters of the various layers of composite resonator and hence the sensitivity of device in terms of these parameters.

An other advantage of the present EQCM theory is that only the characteristic impedance and the propagation constant of lines, in terms of physical parameters of each layer, are necessary for theoretical formulation by means of impedance transformation equation. This formulation greatly simplifies wave scattering calculations of EQCM, or of multilayered structures in the one-dimensional theory and may be generalized for propagation in an arbitrary crystal direction. On the other hand, a structure supporting $N$ propagation modes may also be formally represented as $N$ fictitious transmission lines.

Unlike others EQCM theory, the presented analysis make the formulation and the physical interpretation of various scattering problems easier. It allows simply the prediction of EQCM
with large mass loading in liquid and it facilitates the optimization of electronic of an active EQCM.

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References

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