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Simulating Frequency Nonlinearities in Quartz Resonators at High Temperature and Pressure

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Abstract: In order to facilitate the design of quartz resonators as sensors for the oil and gas industry, the current work focuses on the development of a three-dimensional finite element model to calculate the frequency change of anisotropic quartz resonators associated with the application of temperature and pressure. In doing so, the simulation employs the incremental linear field equations for superimposed small vibrations onto nonlinear thermoelastic stressed media, as given by Lee and Yong [1]. This method involves solving geometric and material nonlinearities for the both the thermal stress and piezoelectric models in COMSOL. The thickness-shear mode frequency response of the model was benchmarked to experimental sensor data with temperature ranging from 50°C to 200°C (in 25°C increments) and pressure from 14 psi to 20,000 psi (with 2,000 psi increments, approximately). The normalized frequency response to the change in external pressure matched very well with experimental data at lower temperatures, and by the same token, the temperature-frequency response matched the experimental trend well for lower pressures. The study found, however, that applying high temperature and pressure simultaneously leads to considerable error in the frequency response. It is hypothesized that the increased error at extreme conditions is due to the current lack of certain material properties of quartz, known as the temperature derivatives of the third-order elastic coefficients.

Keywords: quartz, temperature, pressure, sensor, nonlinear

1. Introduction

Since the early 20th century, quartz sensors have been used to measure such physical parameters as temperature, pressure, and film thickness, among many others. Because of their compact size and rugged characteristics, combined with excellent sensitivity and long-term stability, specialized quartz crystal resonators are well suited to serve as down-hole temperature and pressure sensors in oil and gas wells, a basic example of which is given in Figure 1. Such resonators utilize the nonlinearities in quartz's frequency response associated with changes in temperature and pressure. In spite of the fact that quartz has been used in this regard for the majority of the last century, its complete capability has not been fully realized. The primary reason is because of the complex anisotropic and nonlinear nature of quartz's electro-mechanical properties. In developing an accurate finite element model for the frequency behavior of quartz with respect to external factors, which is the goal of the current work, one must explicitly take into account these complications. Successfully doing so will establish a valuable tool to efficiently explore new sensor designs and additionally yield insight into the modeling of nonlinear multi-physical phenomena in general.

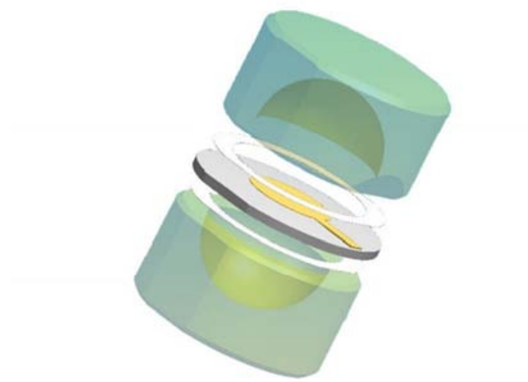

Figure 1: Quartz pressure sensor geometry with end caps [2]

Predicting the frequency response of quartz to changes in temperature and pressure is not a new concept. Empirical data documenting the frequency-temperature behavior of specific cuts was compiled by such researchers as Bechmann et al. in 1962 [3], while similar empirical data for frequency-pressure or frequency-stress response includes the work of EerNisse [4], among many others. Yet two factors preclude this data from direct usage in the current study: the nonlinearity of the *combined* temperature and pressure response which on the whole is much less documented, and the desire to predict the response of previously untested cut angles and geometries of quartz. For this reason, a full finite element solution for the frequency response, which COMSOL provides, is necessary as it is built upon the fundamental anisotropic constitutive properties of quartz, rather than empirical trends. Patel provides such a finite element solution [5], but does not examine the effect of external pressures on frequency response.

This fundamental approach for such a complex material model is made possible by a modification to the standard thermoelastic governing equations, as suggested by Lee and Yong [1]. Their field equations for small vibrations superposed on nonlinear initial strain, referred to here as the incremental method, forms the backbone of the current simulation algorithm. The incremental method allows the full material definition of quartz, including its nonlinear elastic response and temperature dependant properties, to serve as the basis for the returned frequency response. The current work shows that the error of the predicted frequency is highest at the most extreme combined temperature and pressure, likely due to certain material properties of quartz that are not currently available in literature, known as the temperature derivatives of third-order elastic coefficients.

2. The Governing Equations of Piezoelectricity and the Incremental Method

In this context, the incremental method can be seen as a deviation from the governing equations of linear piezoelectricity in stress-charge form, where T_{ij} is the second Piola-Kirchhoff stress tensor, S_{kl} is the Lagrangian strain tensor, D_i is electric displacement, E_j is the electric field, C^E_{ijkl} is stiffness, e_{kij} is the piezoelectric coupling matrix, and ϵ^S_{ij} is dielectric permittivity:

$$T_{ij} = C^E_{ijkl}S_{kl} - e_{kij}E_k$$

$$D_i = e_{ikl}S_{kl} + \epsilon^S_{ij}E_j$$

In the standard COMSOL piezoelectric interface, these linear constitutive equations are combined with the partial differential equations over the domain for mechanical motion and Gauss's Law for an insulator, respectively, where additionally p_0 is density and U_i is displacement ($\ddot{U}_i$ being acceleration):

$$p_0\ddot{U}_i = T_{ij,j}$$

$$D_{i,i} = 0$$

While these equations serve as the most simplistic representation of piezoelectric phenomena, they are not themselves adequate to accurately represent the changes in a crystal oscillator's resonant frequency with temperature and pressure. These trends require temperature dependant material properties and a nonlinear elastic formulation. The incremental method provides these additional features while also allowing the simplification that the piezoelectric vibrations themselves are still linear. This is achieved by dividing the problem into two separate phases connecting three distinct states (rather than one phase connecting two states) as shown in Figure 2.

Namely, the total displacement ($\overline{U_i}$) as referenced from the natural state (stress-free at 25°C) is divided into two parts, the initial displacement (U_i) and the incremental displacement (u_i):

$$\overline{U_i} = U_i + u_i$$

Similarly, total strain and total stress also have both initial and incremental parts:

$$\overline{S_{ij}} = S_{ij} + s_{ij}$$

$$\overline{T_{ij}} = T_{ij} + t_{ij}$$

The initial displacement relates the natural state to the initial state, and the incremental displacement relates the initial state to the final state.

The initial displacement, strain, and stress include the effects of thermal strains and external tractions, and are represented assuming large deformations and nonlinear elasticity. The initial state does not include any piezoelectric effects, and is in that regard, a more traditional solid mechanics formulation. Thus, the constitutive equations for the initial state are that of the standard nonlinear Lagrangian formulation from the theory of elasticity, as follows from Lee and

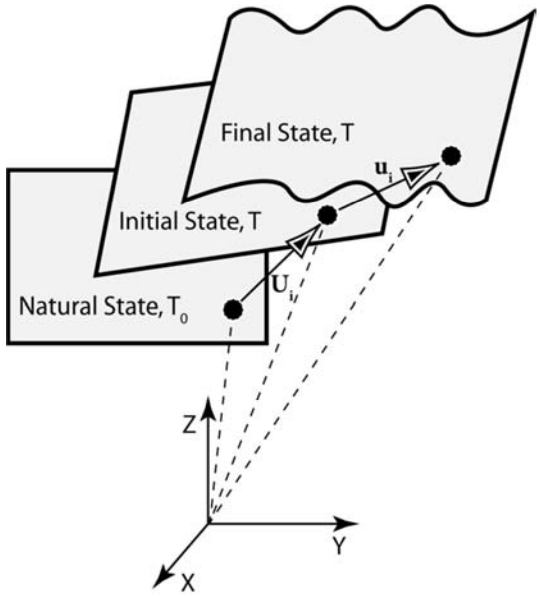

Figure 2: The three states of the incremental method [1]

Yong [1], except that the degree of nonlinearity has been limited to only the lowest two degrees, as these are all that are defined for quartz.

Large deformation strain-displacement:

$$S_{ij} = \frac{1}{2}\left(U_{j,i} + U_{i,j} + U_{k,i}U_{k,j}\right)$$

Nonlinear stress-strain:

$$T_{ij} = C^{\theta}_{ijkl}S_{kl} + \frac{1}{2}C^{\theta}_{ijklmn}S_{kl}S_{mn} - \lambda^{\theta}_{ij}$$

Stress equation of motion:

$$p_0\ddot{U}_i = \left(T_{ij} + T_{jk}U_{i,k}\right)_{,j} \text{ in V}$$

Surface traction equilibrium:

$$P_i = n_j\left(T_{ij} + T_{jk}U_{i,k}\right) \text{ on S}$$

Here, U_i, S_{ij}, T_{ij}, and ρ_0 are as previously described, C^{θ}_{ijkl} and C^{θ}_{ijklmn} are the second- and third-order elastic stiffnesses (respectively), λ^{θ}_{ij} is the stress coefficient of temperature, P_i is the traction on a boundary surface with unit normal vector n_j, and the superscript θ denotes material properties that are functions of temperature. “In V” alludes to the fact that the partial differential equation is to be solved over the volume of the domain, while “on S” similarly corresponds to the boundary surface. It should be noted here that while C^{θ}_{ijklmn} is theoretically a function of temperature, the nature of this function (or equivalently, the “temperature derivative” of the third-order elastic coefficients) is unknown. Therefore in the current finite element algorithm, the third-order elastic coefficients are modeled as temperature independent.

The final state is then governed by the superposition of the incremental displacement onto this initial state. However, the incremental quantities are governed by their own special equations. With the thermal strains and external tractions having been resolved in the initial state, the incremental displacement, strain, and stress take only the effect of piezoelectric vibrations. These vibrations can safely be assumed to be small deformations, and so the incremental equations are made linear during their derivation. (See Lee and Yong [1] for details of the derivation, and for full definitions of temperature dependant material properties.)

Incremental strain-displacement:

$$s_{ij} = \frac{1}{2}\left(u_{j,i} + u_{i,j} + U_{k,j}u_{k,i} + U_{k,i}u_{k,j}\right)$$

Incremental piezoelectricity, stress-charge form:

$$t_{ij} = (C^{\theta}_{ijkl} + C^{\theta}_{ijklmn}S_{mn})s_{kl} - e^{\theta}_{kij}E_k$$

$$D_i = e^{\theta}_{ikl}s_{kl} + \epsilon^{\theta}_{ij}E_j$$

Incremental equation of motion:

$$p_0\ddot{u}_i = \left(t_{ij} + t_{jk}U_{i,k} + T_{jk}u_{i,k}\right)_{,j} \text{ in V}$$

Incremental surface traction equilibrium:

$$p_i = n_j\left(t_{ij} + t_{jk}U_{i,k} + T_{jk}u_{i,k}\right) \text{ on S}$$

Gauss’s Law for an insulator:

$$D_{i,i} = 0 \text{ in V}$$

Note that three *initial* terms $U_{k,j}$, S_{mn}, and T_{jk} (the initial displacement derivative, initial strain, and initial stress, respectively) appear in the *incremental* equations. This is paramount for designing a finite element algorithm around the incremental method for two reasons. First, the presence of these three terms dictates that the initial state must be completely solved *before* attempting to define the final state via the incremental equations. Taking this sequence into account, the three initial terms can therefore be seen as field variables with known values at each point in the continuum when the incremental equations are solved. Secondly, the terms and equations themselves denote exactly *how* those initial results factor into the incremental model.

3. Use of COMSOL Multiphysics

Knowing the theoretical framework provided by the incremental method previously described, a corresponding algorithm within the COMSOL Multiphysics environment is developed. COMSOL is uniquely suited to perform this task for several reasons. First, COMSOL provides in its default form simplified building blocks for

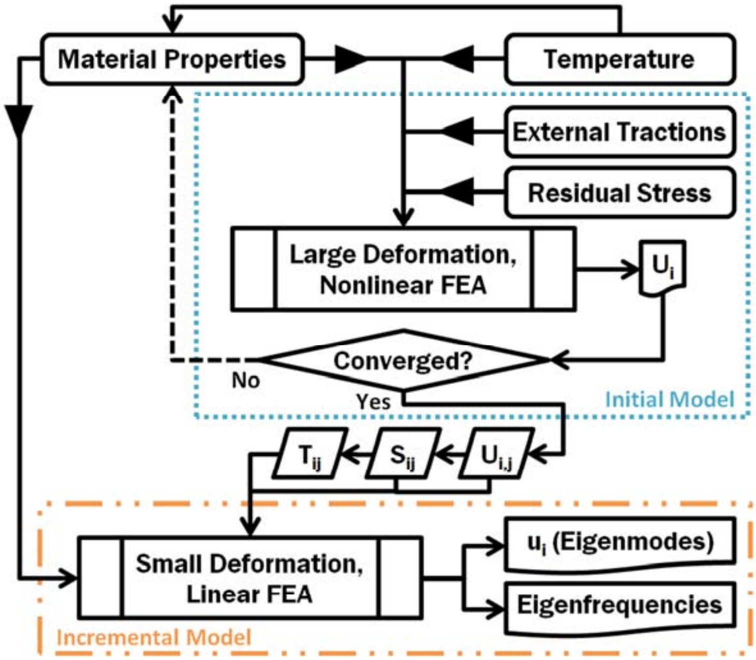

Figure 3: Nonlinear Stressed Homogeneous Temperature (NSHT) Algorithm

constructing the incremental method, being the solid mechanics and piezoelectric interfaces. Furthermore, COMSOL allows those fundamental configurations to be examined and changed, allowing the user to bring in the additional complexities that the incremental method requires. Lastly, the COMSOL environment provides not only finite element models, but the post-processing and global scope to tie these models together into a single, unified algorithm that encompasses the entire incremental method from start to finish.

Knowing this, one can compare the initial equations to the incremental equations and deduce that an algorithm for the complete incremental method must include two separate finite element models, the initial model and the incremental model, each with its own governing expressions. The initial governing equations very closely resemble those of the Thermal Stress interface, while the incremental equations most directly correspond to the Piezoelectric Solid interface. Using these defaults as a template, each is modified to reflect the governing equations previously described, and the result is referred to as the Nonlinear Stressed Homogeneous Temperature (NSHT) Algorithm depicted in Figure 3. Although a full time-dependent simulation would be supported by these same equations, this study is only concerned with predicting the resonant frequency of the piezoelectric vibrations in the oscillator, and thus an eigenfrequency solution of the incremental model is most computationally efficient.

Note that the algorithm is nonlinear in a finite element sense because of the nonlinear elasticity equations in the initial model. COMSOL automatically accounts for this nonlinearity and iterates for convergence accordingly, as shown in Figure 3. Some simplifications can be made on this most general scheme if, for instance, thermal strains are much higher in magnitude than elastic strains, or if the resonator is in a state of homogeneous stress-free thermal expansion. These simplifications are not applicable for the combined high temperature, high pressure case however, and so are not expounded further.

4. Results

After satisfactorily benchmarking the Nonlinear Stressed Homogeneous Temperature (NSHT) Algorithm to various experimental frequency-temperature and frequency-stress trends, (the former including Bechmann et al. [3] and the latter including EerNisse [4]), the algorithm was used to solve for the temperature- and pressure-dependent resonance of a commercially available quartz pressure sensor, shown in Figure 1. This sensor has the geometry of a cylindrical casing surrounding a centrally located circular resonator. The casing, formed by two hermetically sealed end caps, mechanically serves to actuate the state of hydrostatic pressure on the exterior as a non-uniform biaxial compressive stress across the interior AT-Cut resonator plate. This stress measurably influences the resonant frequency of the resonator, and thus external pressure can be assumed from a given frequency.

Although nominally referred to as a "pressure" sensor, such a device does have a considerable temperature response and is thus technically a temperature-pressure sensor. Therefore to measure both in a down-hole application, an independent temperature measurement must also be made. It is likewise imperative that the algorithm be able to output both the pressure and temperature response of the "pressure" sensor, as this allows the simulation to predict how easily the two responses can be separated for a given pressure sensor design.

After the simulation was ran for temperature ranging from 50°C to 200°C and pressure from 14 psi to 20,000 psi, the results from the NSHT Algorithm were then compared to two sets of actual experimental baseline frequency data for the same sensor design. Such a data set includes one dependent variable (relative frequency in parts per million) as a function of two independent variables (temperature and pressure). While this data would be most directly conveyed as a 3D surface or carpet plot, such a representation is not desirable for comparing the algorithm against the experimental baseline. One alternative is to take 2D slices of these surfaces that can be of either constant temperature (isothermal) or constant pressure (isobaric). The former approach results in the plots shown in Figure 4 and Figure 5. Note that frequency data is normalized to the frequency at 50°C and 14 psi for the calculation of frequency shift in parts per million.

Correlation between the experimental data and the simulation is fairly good overall, with the major experimental trend being mirrored in the

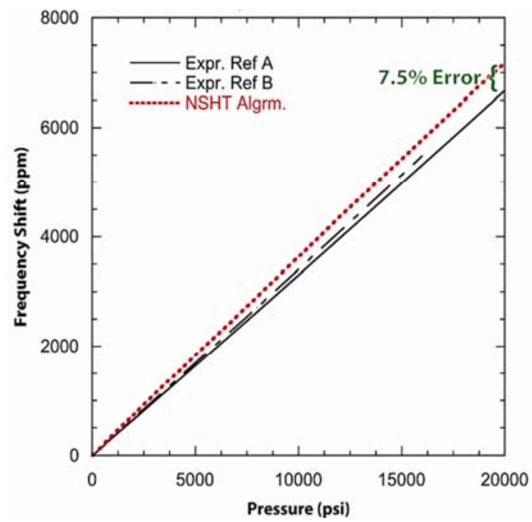

Figure 4: Isothermal simulation results; T = 50°C

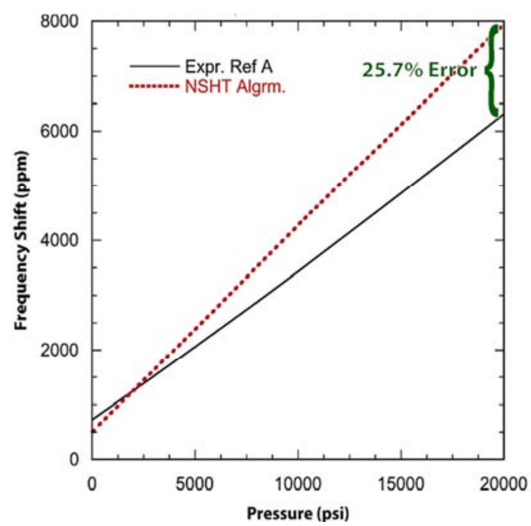

Figure 5: Isothermal simulation results; T = 200°C

output of the algorithm. For the isothermal plots the pressure response is linear, and the slope of the line in the simulation is near the experimental slope, especially at low temperatures. At higher temperatures however, the experimental slope of the pressure response decreases while the simulation's slope increases slightly. This results in the simulation's error compared to experimental values growing from 7.52% at 50°C and 20,000 psi to 25.65% at 200°C and 20,000 psi, as noted in Figure 4 and Figure 5.

5. Conclusions

It is this second-order experimental change in the first-order trend, like the decreasing slope of the pressure response with increasing temperature, where the current model shows little correlation. It is known that the third-order elastic coefficients of stiffness are the gateway to the frequency shifts due to a stress bias [6]. In other words, the nonlinear elastic terms control the pressure response of frequency. Therefore, in order for the pressure response to change accurately with temperature, the inputted third-order elastic coefficients also need to change accurately with temperature in the material definition. The accurate change of the third-order elastic coefficients is, of course, given by their temperature derivatives. Unfortunately, these temperature derivatives are not currently known. This leads to the nonlinear elasticity terms being modeled as constants, which in turn generates inaccuracies in pressure response as the temperature is increased away from the reference temperature of 25°C.

In all, the developed NSHT Algorithm for applying the incremental method within COMSOL is shown to provide some degree of accuracy for predicting the frequency response of quartz pressure sensors, especially around either ambient temperature or ambient pressure. Furthermore, seeing the potential for the possible benefit of implementing the third-order elastic coefficients as functions of temperature should aid future researchers in deciding if defining their full anisotropic temperature derivatives is a worthwhile undertaking. Doing so would definitely make the current model more acute to some degree, and improve the simulation's ability to meet the complex design challenges posed for quartz sensors in extreme environments.

6. References

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7. Acknowledgement

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