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IDENTIFICATION OF THE PROPERTIES OF A COMPLEX LAYER DEPOSITED ON THE SURFACE OF A QUARTZ CRYSTAL MICROBALANCE (QCM) BY THE IMPEDANCE MEASUREMENT

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Abstract - The quartz crystal microbalance (QCM) is a quartz crystal plate resonator for measuring a minute mass in terms of the resonant frequency change. In some applications, an adsorbing layer must be formed on the resonator surface, which adsorbs the material whose mass is measured. This layer affects not only the resonant frequency but also its damping as it is viscoelastic. Its presence can not simply be ignored but should be included in the modeling. In our previous work, the algorithm to characterize the viscoelastic layer's properties was developed, in which the multiple resonant frequencies and the corresponding resonant resistances were considered including the overtone operation of the quartz crystal plate [1]. It is unrealistic however to assume that the layer's properties are unchanged for such a wide frequency range. In the present paper, no overtone resonance is considered. Mass of the adsorbed material and the thickness of the viscoelastic layer are identified by means of the Newton's method.

1. INTRODUCTION

Quartz Crystal Microbalance, abbreviated to QCM, is a plate resonator made of quartz crystal operated in thickness shear mode. A thin mass layer deposited on the surface of the QCM can be measured in terms of the decrease of the resonant frequency. The ratio of the loading mass to the equivalent mass of the plate resonator is proportional to the frequency change against the resonant frequency. The frequency is the quantity most accurately measured, of the order of $\Delta f/f_0 = 10^{-7\sim-8}$. This corresponds to the mass of the order of nano-gram in the case of standard QCMs of 9MHz. The QCM is widely used, such as, for vacuum vapor deposition monitoring. Application is also extended to the characterization of biomedical materials due to its extreme sensitive capability, in which the layer is selectively formed over a certain layer provided on the surface of the QCM by the physical adsorption or chemical reaction. The operation should also be assumed to the operation under watery atmosphere [2]. The viscoelastic layer that traps a material to be measured is not always solid but sometimes viscoelastic [3]. It can be a lossy material and its stiffness can be much smaller than that of the quartz crystal, of the order of one-tenth. Under this circumstance, the more elaborate model and data must be considered. We again introduce a distributed parameter model, which is solved for the impedance data measured at the electrical terminals, where the thickness of the adsorbing layer and the mass of the adsorbed material are to be identified. The multiple root finding technique based on the Newton's method is utilized for parameter searching.

2. MODELS

Several types of modeling are possible for the analysis of the QCM responses. Two types are shown in Figures 1 and 2.

Classical (lumped parameter) model Figure 1(a) shows a lumped parameter model. The model is reasonable when the layer thickness is much shorter than the wavelength of the excitation, and the loading layer moves totally in phase. The additional mass is identified due to the resonant frequency shift. The resonant frequency is given as

$$f_m = \frac{1}{2\pi}\sqrt{\frac{K}{M+\Delta m}} = \frac{1}{2\pi}\sqrt{\frac{K}{M(1+\Delta m/M)}} = \frac{1}{2\pi}\sqrt{\frac{K}{M}}\left(1+\frac{\Delta m}{M}\right)^{-\frac{1}{2}} \approx f_0\left(1-\frac{1}{2}\frac{\Delta m}{M}\right) \tag{1}$$

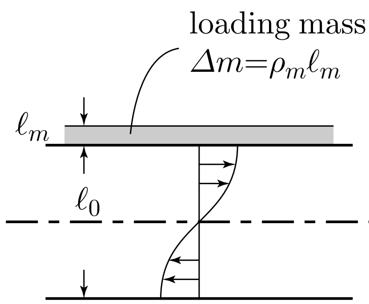
where $f_0 (= 1/(2\pi)\sqrt{K/M})$ is the resonant frequency of the plate resonator without mass loading, and Maclaurin expansion is used for the approximation. The equivalent mass M is a half of total mass M_0 of the plate for the lowest mode. The normalized frequency shift $\Delta f/f_0$ is

$$\frac{\Delta f}{f_0} \simeq -\frac{1}{2}\frac{\Delta m}{M} = -\frac{\Delta m}{M_0} \tag{2}$$

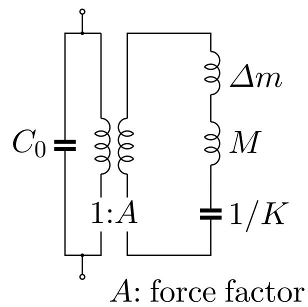
When the frequency shift Δf is measured, the mass is straightforwardly determined by

$$\Delta m = -\frac{\Delta f}{f_0} M_0 \tag{3}$$

ρ_m and ℓ_m as shown in Figure 1(a), are mass density and thickness of the additional layer. Figure 1(b) shows an equivalent circuit representation, made of damped (shunt) capacitance, equivalent mass and stiffness of the plate resonator, C_0, M and K. Since the dissipation in the quartz crystal plate is so small as to be negligible. Mechanical loss or damping in adsorbed mass can not be considered in this model.



(a) Physical model



(b) Equivalent circuit

Figure 1. Classical (Lumped parameter) model.

Transmission-line (distributed parameter) model Figure 2 shows a distributed parameter model. This model is utilized in the present paper. The viscoelastic layer deposited on the resonator surface can also be modeled with a distributed lossy transmission line on the top surface of which a mass loading is placed. The viscoelastic layer is not uniformly moving when the QCM is operating. In Figure 2(b), Z_R is the impedance, which is looked toward the deposited layer-mass loading system. That is given as

$$Z_R = z_m \frac{j\omega m_s + z_m \tanh(\gamma_m \ell_m)}{z_m + j\omega m_s \tanh(\gamma_m \ell_m)} \tag{4}$$

where z_m, ℓ_m, m_s and ω are the characteristic impedance, the thickness of the viscoelastic layer, mass loading adsorbed on the viscoelastic layer and angular frequency, respectively. γ_m is the propagation constant, which is defined by the transmission-line theory [4] as

$$\gamma_m = \frac{j\omega\sqrt{\rho_m}}{\sqrt{G_m + j\omega\,\eta_m}} \tag{5}$$

where ρ_m, G_m and η_m are mass density, shear modulus and viscosity of the layer, respectively. Z_{in} is the input impedance on the mechanical terminals in the equivalent circuit of the resonator as

$$Z_{in} = z_0 \frac{Z_R + z_0 \tanh(\gamma_0 \ell_0)}{z_0 + Z_R \tanh(\gamma_0 \ell_0)} \tag{6}$$

where z_0, γ_0, ℓ_0 are the characteristic impedance, the propagation constant and the thickness of the resonator plate, respectively. This impedance Z_{in} is in reality measured in terms of the electric impedance at the electrical terminals. The resonant frequency can be determined as the frequency at which the imaginary part of the input impedance Z_{in} is zero. It is difficult to solve analytically for the resonant frequency and resonant resistance. The solution could numerically be made using optimization technique such as the Newton's method.

When ℓ_m is very small, Z_R is approximated to be

$$Z_R = j\omega\,(m_s + m_m) \tag{7}$$

where m_m is the mass of the layer ($m_m = \rho_m \ell_m$). This is equivalent to the case when the lumped mass loading $m_s + m_m$ is provided on the surface.

When no dissipation is considered and $m_s + m_m$ is small, solving eqn.(6), for $Z_{in} = 0$, the solution is,

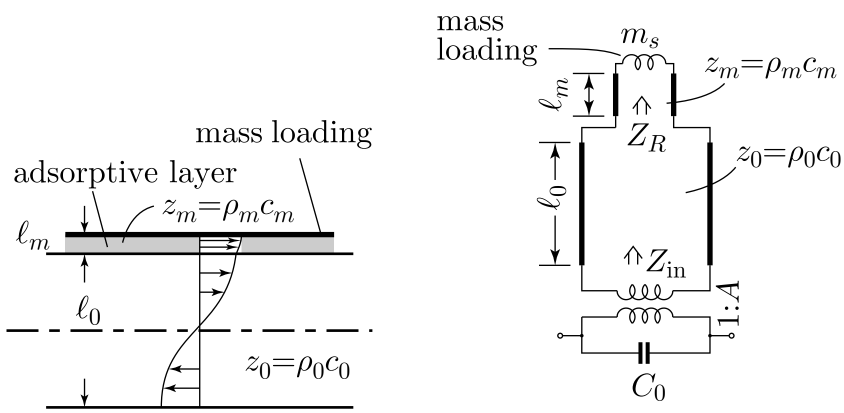
$$\omega_m = \frac{\pi z_0}{M_0 + m_s + m_m} \tag{8}$$

where ω_m is the resonant frequency solution when mass loading $m_s + m_m$ is provided. Normalized frequency shift in this case is given as

$$\frac{\Delta f}{f_0} = \frac{\Delta\omega}{\omega_0} = \frac{\omega_m - \omega_0}{\omega_0} = -\frac{m_s + m_m}{M_0 + m_s + m_m} = -\frac{\Delta m}{M_0 + \Delta m} \simeq -\frac{\Delta m}{M_0} \tag{9}$$

which is the same as the case of our classical lumped parameter model.

It should be noted that the transmission-line model leads the same result as that of the classical model when the viscoelastic layer is very thin.



(a) Physical model

(b) Equivalent circuit

Figure 2. Transmission-line (distributed parameter) model.

3. FORWARD ANALYSIS

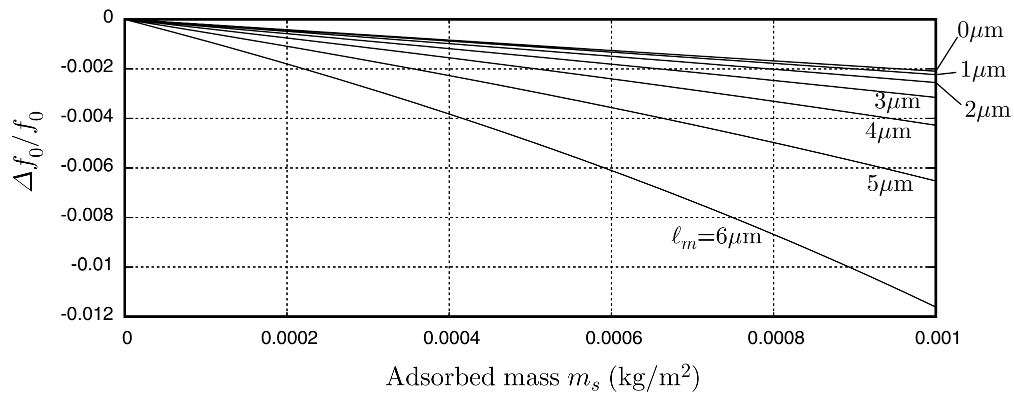
We consider the case when the thickness of the layer is as thick as one quarter wavelength. Physical parameters used in the present numerical simulation are shown in Table 1, where quality factor Q is defined as $Q = G/\eta$, where η is the damping factor. The physical parameters G_m, ρ_m and η_m of the viscoelastic layer are known, while ℓ_m and m_s are to be determined from the measurement of the resonant frequency and the resonant resistance.

Table 1. Physical parameters assumed.

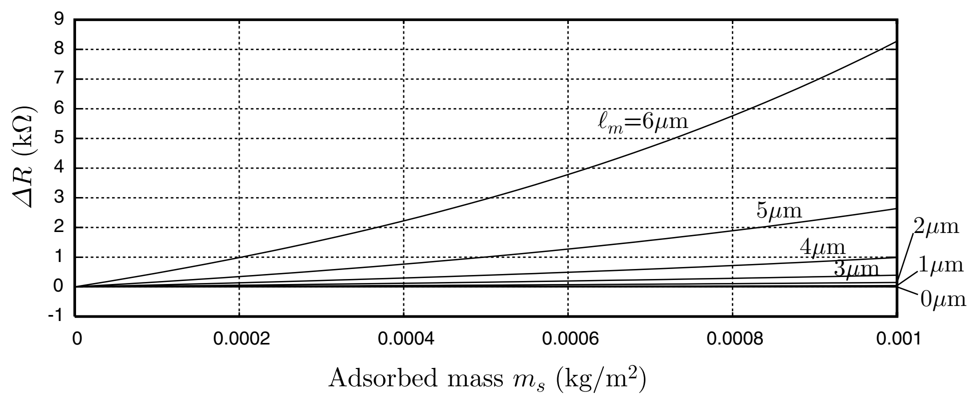
	Mass density ρ [kg/m^3]	Shear modulus G [Pa]	Quality factor Q	Thickness ℓ [m]	Wavelength λ [m] for 9MHz
Resonator	$\rho_0 : 2650$	$G_0 : 2.87 \times 10^{10}$	$Q_0 : 1 \times 10^5$	$\ell_0 : 0.18 \times 10^{-3}$	$\lambda_0 : 0.36 \times 10^{-3}$
Adsorbing layer	$\rho_m : 1000$	$G_m : 1 \times 10^8$	$Q_m : 1 \times 10^2$	$\ell_m : 0 \sim 6 \times 10^{-6}$	$\lambda_m : 35 \times 10^{-6}$

The resonant frequency shift $\Delta f_0 = f_0 - f_0|_{m_s=0}$ and resonant resistance shift $\Delta R = R - R|_{m_s=0}$ curves are shown in Figure 3 for the layer thickness ℓ_m from $0\,\mu$m to $6\,\mu$m.

Figure 3 shows that both $\Delta f_0/f_0$ and ΔR depend on ℓ_m, the thickness of the adsorbing layer. The sensitivities, the gradients of $\Delta f_0/f_0$ and ΔR become higher as the thickness of the adsorbing layer increases, and the curves are both not straight.



(a) f_0 shift versus adsorbed mass m_s



(b) R shift versus adsorbed mass m_s

Figure 3. Forward analysis.

4. PARAMETER ESTIMATION

In the actual application, Δf_0 and ΔR are both given by the measurement. The unknown parameters are the thickness of the layer ℓ_m and the adsorbed mass m_s. If it is assumed that Δf_0 and ΔR change independently, the parameters ℓ_m and m_s can be determined from the measured values of Δf_0 and ΔR.

Figure 4 shows the relation of m_s to ℓ_m, when both resonant frequency shift and the resonant resistance shift are known. Suppose we have obtained the measured values for $\Delta f_0 = -1\,\mathrm{kHz}$ and $\Delta R = 0.2\,\mathrm{k\Omega}$. The unknown values ℓ_m and m_s are uniquely determined from measured values Δf_0 and ΔR at least within the plotted range. From the crossing point, we have the solutions, $\ell_m \simeq 4\,\mu\mathrm{m}$ and $m_s \simeq 3 \times 10^{-4}\,\mathrm{kg/m^2}$.

To obtain the solutions, in reality, these crossing points should quickly be searched, which is made here by using the multiple root finding algorithm based on the simple Newton's method. This algorithm is used for solving the equation, $\boldsymbol{y}(\boldsymbol{x}) = \boldsymbol{0}$, where $\boldsymbol{y}(\boldsymbol{x})$ is a vector function of a parameter vector $\boldsymbol{x}$.

In the present case, the vector function $\boldsymbol{y}$ consists of error functions so that

$$\boldsymbol{y} = \{f_{\mathrm{err}}, R_{\mathrm{err}}\}^T \tag{10}$$

where error functions f_{err} and R_{err} are chosen to be

$$\begin{aligned} f_{\mathrm{err}} &= \Delta f_0 - \Delta \hat{f}_0 \\ R_{\mathrm{err}} &= \Delta R - \Delta \hat{R} \end{aligned} \tag{11}$$

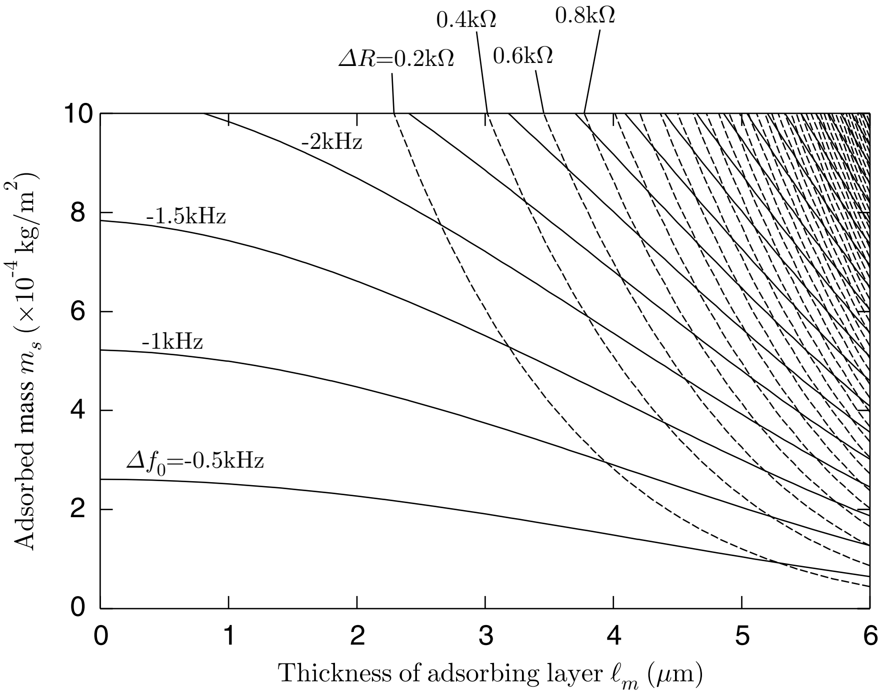


Figure 4. The relation between the thickness of the adsorbing layer ℓ_m and the adsorbed mass m_s for the resonant frequency shift Δf_0 and resonant resistance shift ΔR.

The values with a hat ($\hat{\ }$) indicate the exact values. The parameter vector $\boldsymbol{x}$ is taken to be

$$\boldsymbol{x} = \{\ell_m, m_s\}^T \tag{12}$$

The iteration is complete when $||\boldsymbol{y}||$ becomes small enough.

In the simulation, forward solutions are used for the "measured" values. The values of ℓ_m and m_s are assumed to be $3\,\mu$m and $5\times10^{-4}\,$kg/m^2 respectively. The initial values are set to $\{\ell_m, m_s\}^T = \{2,\ 3\times10^{-4}\}^T$. The solution is successfully found after 7 iterations as shown in Figure 5. The solution path is indicated by "Path 1".

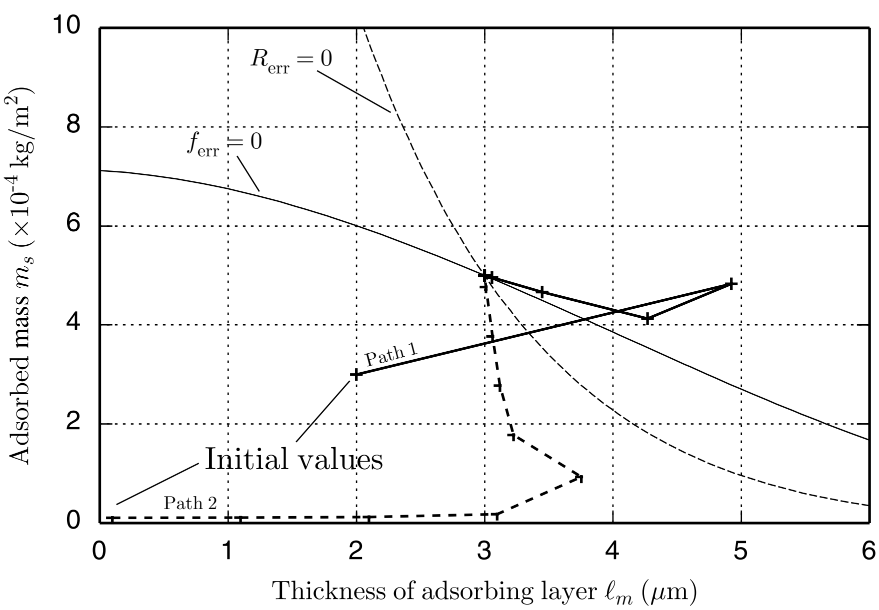


Figure 5. Solution paths.

If smaller initial values are set to be $\{\ell_m, m_s\}^T = \{0.1, 0.1 \times 10^{-4}\}^T$, however, the process is likely to diverge. This is because $f_{\rm err}$ and $R_{\rm err}$ is almost flat in that vicinity. In order to provide more stable search process avoiding such a divergence, limited step size $||\Delta \boldsymbol{x}||$ must be devised. With such a modification, the search process converges toward correct values as "Path 2" shown in the figure. Inverse search is correctly carried out in all cases for the range from 2 to 6 for ℓ_m and from 2 to 10 for m_s.

Figure 6 includes the case when the much higher initial values are set to be $\{\ell_m, m_s\}^T = \{15,\ 1 \times 10^{-4}\}^T$. The combinations of ℓ_m and m_s are shown by '+' signs numbered. Convergences are all completed but come to the false places with corresponding numbers. This is due to the fact that there is another crossing point that satisfies $f_{\rm err} = R_{\rm err} = 0$ in the higher range of the thickness, out of the range shown in Figure 4.

Figure 7 shows the trace of a case in which two pattern of the convergence paths are shown. With the initial values $\{\ell_m, m_s\}^T = \{0.1,\ 0.1 \times 10^{-4}\}^T$, path 1 successfully approaches to the true values, while path 2, starting with another initial values $\{\ell_m, m_s\}^T = \{15,\ 1 \times 10^{-4}\}^T$, approaches to the another values $\{\ell_m, m_s\}^T = \{17.3,\ 0.268 \times 10^{-4}\}^T$. The second one belongs to another higher mode of resonances. The areas corresponding to valid and invalid initial value combinations are shown in the same figure. Here, "Valid" means initial values that leads to true values, and "Invalid" means ones that leads to incorrect values. According to this figure, smaller initial values may lead to correct true values as long as it does not diverge.

5. CONCLUSIONS

A QCM system is modeled with a transmission-line (distributed parameter) model, in which the effect of the viscoelasticity of the adsorbing layer provided on the plate surface is included. The mass of the material adsorbed on the viscoelastic adsorbing layer is identified simultaneously with the layer's thickness by means of a simple Newton's method. In the simulation, the forward analysis solution is used for the "measured" data, which are the resonant frequency shift Δf_0 and the resonant resistance shift ΔR for the various thickness of the adsorbing layer ℓ_m and the various adsorbed mass m_s.

In reality, ℓ_m and m_s are to be estimated from the "measured" values of Δf_0 and ΔR. This inverse process was made using the multiple root finding technique based on the Newton's method. The estimation is successfully carried out provided that a proper choice of the initial values are made. With inadequate initial values, the estimation process may converge to incorrect values, which also satisfies the conditions of the error functions $f_{\rm err} = R_{\rm err} = 0$.

REFERENCES

1. N. Wakatsuki, S. Wada, Y. Kagawa and M. Haba, Determination of the mass density of the layer deposited on the surface of the resonator in QCM(Quartz Crystal Microbalance), *Inverse Problems in Engineering Mechanics IV*, (ed. M. Tanaka), 2003, pp.121–126.

2. Z. Lin, C. M. Yip, I. S. Joseph and M. D. Ward, Operation of an ultrasensitive 30-MHz quartz crystal microbalance in liquids. *Anal. Chem.* (1993) **65**, 1546–1551.

3. URL: `http://www.q-sense.com/qcmd_tech.html`

4. J. D. Ryder, *Networks, Lines and Fields*, 2nd edn., Maruzen, Tokyo, 1969.

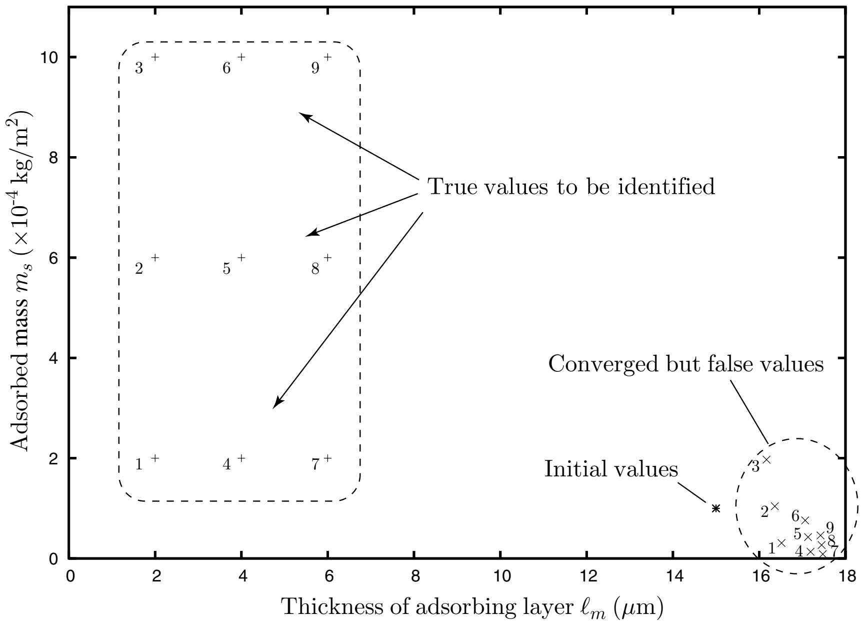


Figure 6. Incorrect convergence.

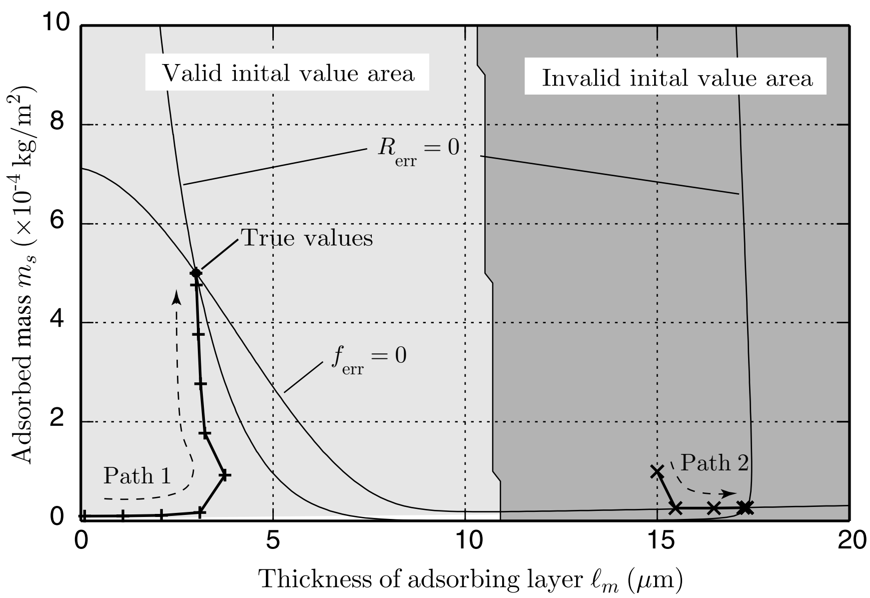


Figure 7. Initial value dependency.