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RESONATOR RESPONSE TO NON-NEWTONIAN FLUIDS

S.J. Martin, T.W. Schneider, G.C. Frye, and R.W. Cernosek
Microsensor Research and Development Department
Sandia National Laboratories, Albuquerque, New Mexico 87185

S.D. Senturia
Department of Electrical Engineering and Computer Science
Massachusetts Institute of Technology, Cambridge, MA 02139

A thickness-shear mode (TSM) resonator typically consists of a thin disk of AT-cut quartz with circular electrodes patterned on both sides (Fig. 1). An RF voltage applied between these electrodes excites a shear mode mechanical resonance when the excitation frequency matches the crystal resonant frequency. When the TSM resonator is operated in contact with a liquid, the shear motion of the surface generates motion in the contacting liquid. The liquid velocity field, $v_x(y)$, can be determined by solving the one-dimensional Navier-Stokes equation [1]:

$$\eta \frac{\partial^2 v_x}{\partial y^2} = \rho \dot{v}_x \tag{1}$$

where ρ and η are the liquid density and shear viscosity, respectively. The solution to Eq. 1 is [1]

$$v_x(y,t) = v_{xo} \exp(-\gamma y) \exp(j\omega t) \tag{2}$$

where v_{xo} is the surface particle velocity, ω is angular frequency and $j = (-1)^{1/2}$. Eq. 2 represents a damped shear wave *radiated* into the contacting liquid by the oscillating resonator surface; γ is a propagation constant for this wave, determined by substituting Eq. (2) into Eq. (1).

Liquid coupling leads to energy storage and power dissipation in the contacting liquid and affects resonator response. An equivalent circuit model can be used to describe both the electrical response of the dry and the liquid-contacted resonator (Fig. 2): a "static" capacitance C_o^* (includes any parasitic capacitance) in parallel with a "motional" branch (L_1, C_1, R_1, L_2, and R_2). The static capacitance arises between electrodes across the insulating quartz. The motional impedance is due to electrical excitation of a shear-mode mechanical resonance in the piezoelectric quartz. Liquid coupling to the surface modifies this motional impedance, introducing a motional inductance (L_2) and a resistance (R_2). Electrical energy storage in L_2 arises from the kinetic energy of the viscously-entrained liquid layer; power dissipation in R_2 arises from the radiation of a damped shear wave into the liquid.

The motional elements, L_2 and R_2, are related to the real and imaginary components of the liquid decay constant γ [2]:

$$R_2 + jX_2 = \frac{nN\pi\eta\gamma}{4K^2\omega_s C_o(\mu_q\rho_q)^{1/2}} \tag{3}$$

where n is the number of sides contacted by liquid, N is the resonator harmonic number, $X_2 = \omega_s L_2$, ω_s is the angular series resonant frequency, where ρ_q, μ_q and K^2 are the quartz density, shear stiffness, and electromechanical coupling factor, respectively.

Newtonian Fluid Loading When the TSM resonator is contacted by a Newtonian fluid, for which viscosity is constant ($\eta \equiv \eta_o$, independent of oscillation amplitude and frequency), Eqs. 1 and 2 yield $\gamma = (\omega\rho/2\eta_o)^{1/2}(1+j)$. This decay constant, with equal real and imaginary parts, leads to a *critically damped* shear wave in the fluid. Substituting this γ into Eq. 3, and separating real and imaginary parts, gives the motional elements arising from *Newtonian fluid* loading [3]:

$$R_2^{(N)} = X_2^{(N)} = \frac{nN\pi}{4K^2\omega_s C_o}\left(\frac{\omega_s \rho \eta_o}{2\mu_q \rho_q}\right)^{\frac{1}{2}} . \tag{4}$$

Eq. 4 indicates that the response of a smooth TSM resonator loaded by a Newtonian fluid depends only on the product ($\rho\eta$) of liquid density and viscosity. Moreover, a distinguishing feature of Newtonian fluid loading is $R_2 = X_2$.

Maxwell Fluid Loading The simplest Non-Newtonian fluid has a viscosity that varies with frequency as

$$\eta(\omega) = \frac{\eta_o}{1 + j\omega\tau} \tag{5}$$

where η_o is the low-frequency viscosity and τ is a shear relaxation time. If the liquid is sheared slowly ($\omega\tau \ll 1$), liquid molecules are able to flow past one another and the fluid behaves as an ideal Newtonian fluid with $\eta = \eta_o$. If sheared rapidly ($\omega\tau \gg 1$), liquid molecules cannot flow and strains are accommodated elastically through molecular deformation. In a Maxwell fluid, the relaxation time $\tau = \eta_o/\mu$, where μ is the high-frequency shear modulus for the liquid. At high frequency, when $\omega\tau \gg 1$, $\eta(\omega) \cong \mu/(j\omega)$, so that the Maxwell fluid behaves as an elastic solid.

When the TSM resonator is contacted by a Maxwell fluid, with viscosity varying according to Eq. 5, the decay constant satisfies $\gamma^2 = (j\omega\rho/\eta_o)(1+j\omega\tau)$. For $\omega\tau \ll 1$, γ matches the Newtonian case above; for $\omega\tau \gg 1$, $\gamma = j\omega(\rho/\mu)^{1/2}$, representing an *undamped* shear wave radiated into an elastic solid. The motional impedance elements arising from *Maxwell fluid* loading are:

$$X_2^{(M)} = X_2^{(N)} \left(1 - \frac{\omega\tau}{\sqrt{1 + (\omega\tau)^2}} \right)^{\frac{1}{2}} \quad \text{(6a)}$$

$$R_2^{(M)} = R_2^{(N)} \left(1 + \frac{\omega\tau}{\sqrt{1 + (\omega\tau)^2}} \right)^{\frac{1}{2}} . \quad \text{(6b)}$$

When $\omega\tau = 0$, the Maxwell fluid behaves as a Newtonian fluid so that $X_2^{(M)} = X_2^{(N)}$ and $R_2^{(M)} = R_2^{(N)}$, as expected. For $\omega\tau > 0$, $R_2^{(M)} > X_2^{(M)}$ and a Non-Newtonian response is indicated. From Eqs. 6, the relaxation parameter $\omega\tau = c/(1 - c^2)^{1/2}$, where $c = (R_2^2 - X_2^2)/(R_2^2 + X_2^2)$.

Fig. 3 shows R_2 and X_2 calculated from both the Newtonian (Eq. 4) and Maxwell (Eqs. 6) fluid models vs. liquid viscosity (η_o) for several values of the liquid shear parameter μ. For the Newtonian fluid, $R_2^{(N)} = X_2^{(N)}$ (dashed line) for all values of viscosity and independent of μ. For the Maxwell fluid, the resistance $R_2^{(M)}$ and reactance $X_2^{(M)}$ converge to the Newtonian fluid values at low values of viscosity, where $\omega\tau \ll 1$. At higher viscosities, where $\omega\tau \geq 1$, the components calculated for Maxwell fluid loading diverge from those due to a Newtonian fluid: $X_2^{(M)}$ falls below the Newtonian prediction, while $R_2^{(M)}$ lies above.

Fig. 4 shows the variation in motional impedance components vs. temperature for a 5 MHz quartz TSM resonator immersed in a polystyrene melt (MW=2,500). Since viscosity varies *inversely* with temperature, the impedance components behave much like the predictions of Fig. 3 for the Maxwell fluid: $R_2 > X_2$, with X_2 exhibiting non-monotonic behavior.

Table I lists the values of R_2 and X_2 obtained from resonator measurements made on several liquids. The glycerol/water mixtures have $R_2 \cong X_2$, giving $\omega\tau \cong 0$ and indicating Newtonian fluid behavior. Paraffin shows a transition from Newtonian to non-Newtonian behavior as temperature decreases (η increases). The polystyrene and polyethylene melts have $R_2 > X_2$ at all temperatures measured, giving a non-zero $\omega\tau$ value and indicating Non-

Newtonian behavior. The data show that the relaxation time τ is only significant for complex molecules. The decrease in τ with temperature is consistent with the Maxwell model ($\tau = \eta_o/\mu$) and can be attributed to a decrease in fluid viscosity η_o.

In conclusion, Newtonian fluids cause an equal increase in resonator motional resistance and reactance, $R_2^{(N)} = X_2^{(N)}$, with the response depending only on the liquid density-viscosity product ($\rho\eta$). Non-Newtonian fluids, as illustrated by the simple example of a Maxwell fluid, can cause unequal increases in motional resistance and reactance. For the Maxwell fluid, $R_2^{(M)} > X_2^{(M)}$, with the relaxation time τ proportional to the difference between $R_2^{(M)}$ and $X_2^{(M)}$. Early results indicate that the TSM resonator can be used to extract properties of non-Newtonian fluids.

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Figure Captions:

Fig. 1. Cross-sectional view of a TSM resonator with the upper surface contacted by a liquid. Shear motion of the surface causes a thin layer of the contacting liquid to be viscously entrained.

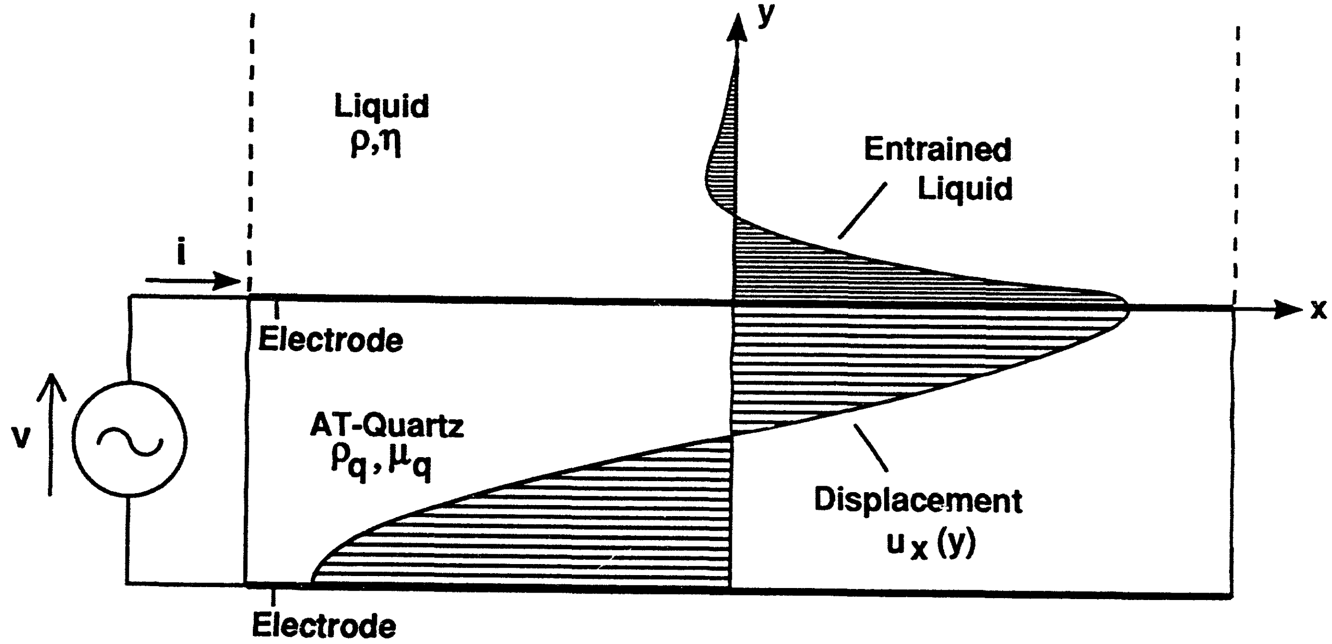
Fig. 2. Equivalent-circuit model to describe the electrical characteristics (for ω near ω_s) of a TSM resonator with liquid loading.

Fig. 3. Motional impedance components R_2 and X_2 calculated from both the Newtonian (Eq. 4) and Maxwell (Eqs. 6) fluid models vs. liquid viscosity (η_o) for several values of the liquid shear parameter μ: (a) 3×10^7, (b) 1×10^8, (c) 3×10^8 dyne/cm^2.

Fig. 4. Motional impedance components vs. temperature for a 5 MHz quartz TSM resonator immersed in a polystyrene melt (MW=2,500).

Table I.

Liquid	Temp. (°C)	R_2 (kΩ)	$X_2 \equiv \omega_s L_2$ (kΩ)	$\omega_s \tau$
H_2O	20	0.341	0.342	≈ 0
43% glycerol in H_2O	20	0.717	0.718	≈ 0
64% glycerol in H_2O	20	1.30	1.30	≈ 0
paraffin	100	0.95	1.00	≈ 0
	60	1.43	1.45	≈ 0
	50	8.12	2.38	1.56
	40	22.4	5.84	1.79
polystyrene	135	17.2	12.8	0.15
	120	28.0	21.0	0.14
	105	45.1	21.8	0.35
polyethylene	300	4.33	1.05	1.94
	200	5.08	0.99	2.47



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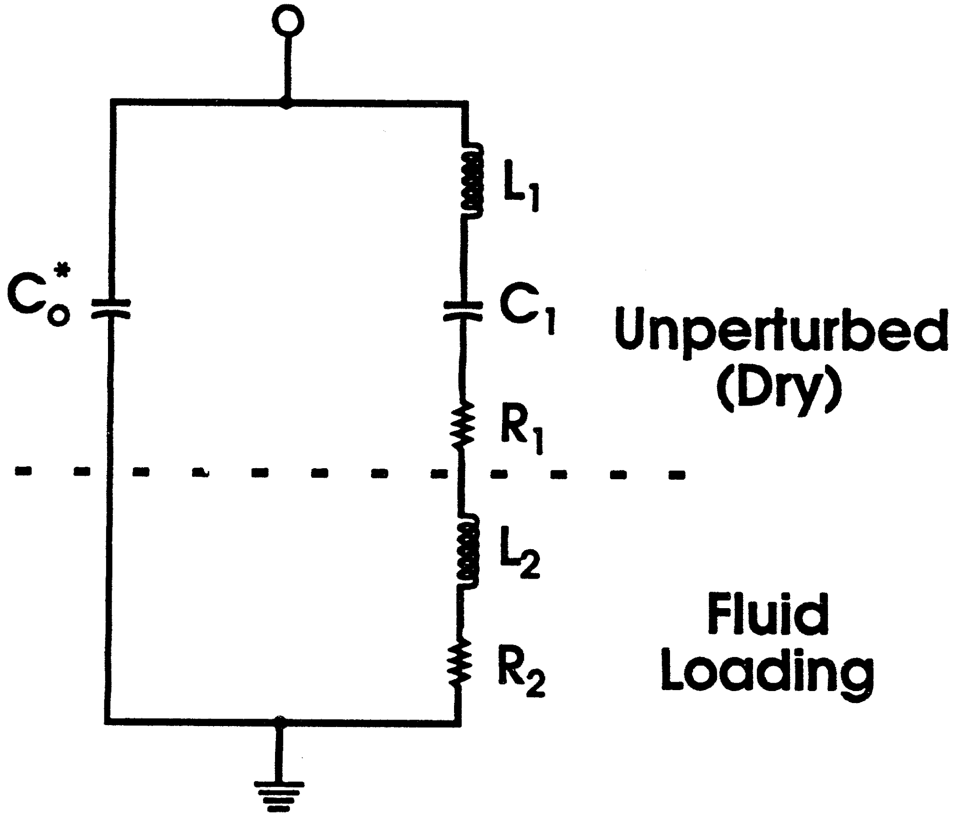

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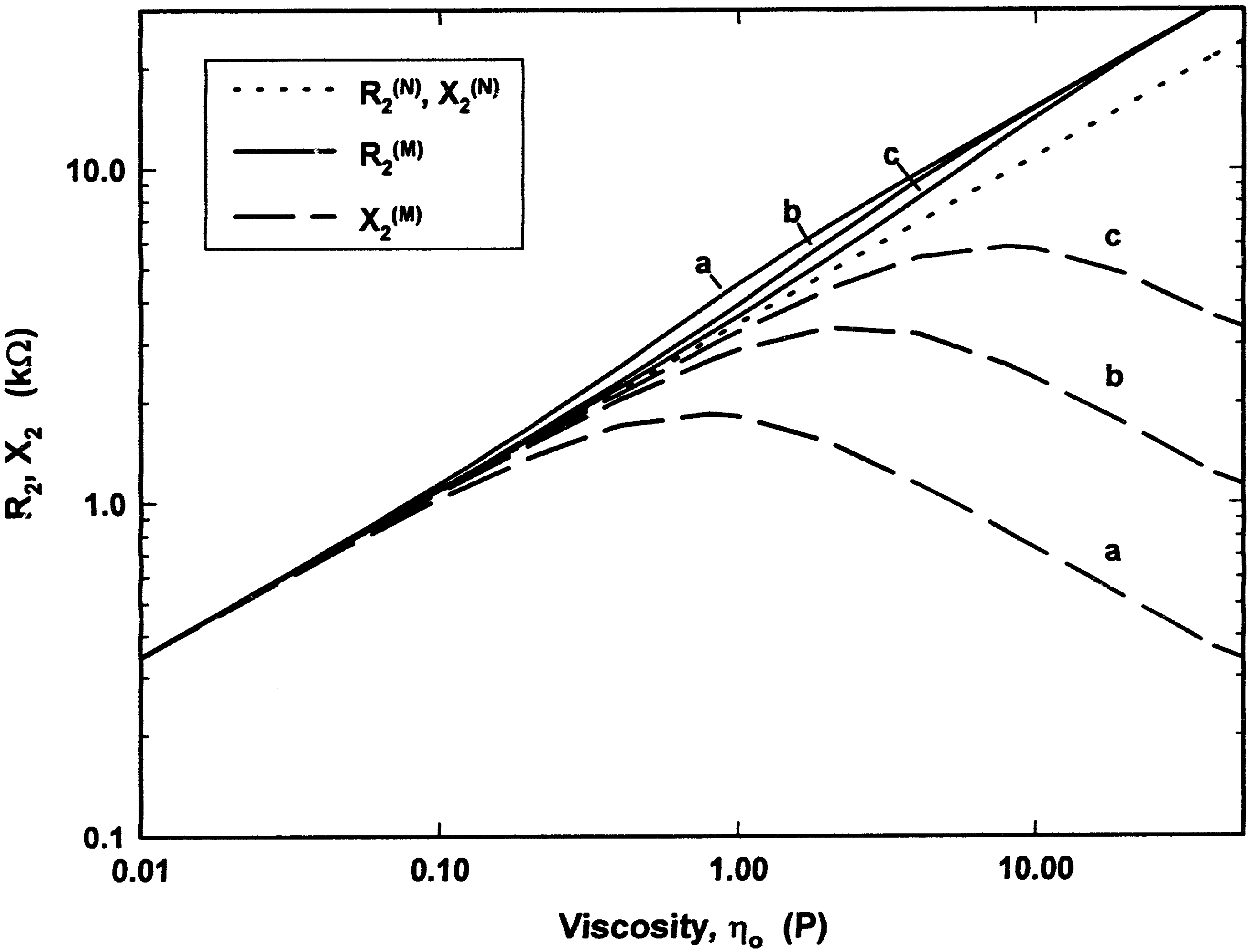

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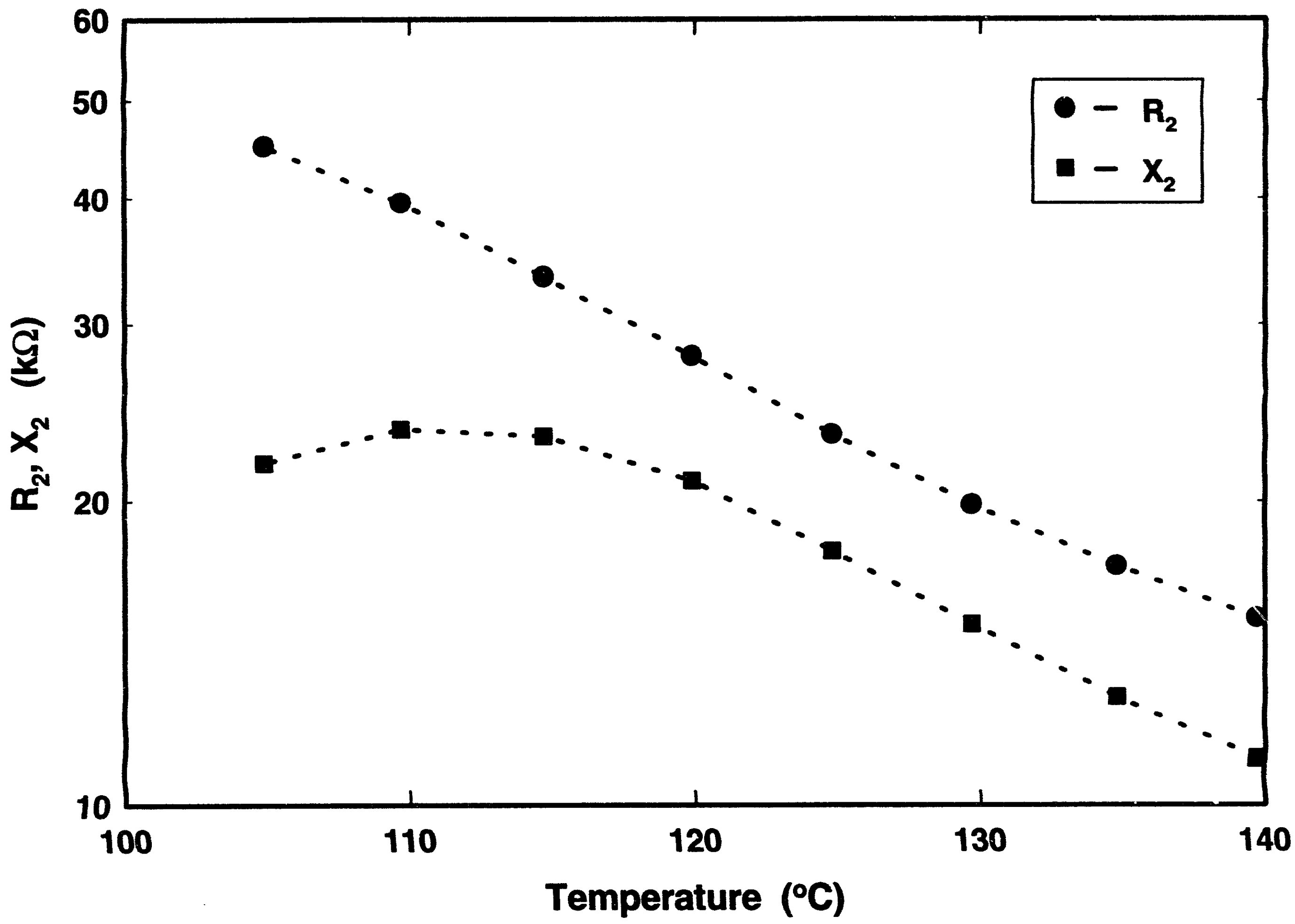

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