Vibration analysis of a new polymer quartz piezoelectric crystal sensor for detecting characteristic materials of volatility liquid*

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We present a new polymer quartz piezoelectric crystal sensor that takes a quartz piezoelectric crystal as the basal material and a nanometer nonmetallic polymer thin film as the surface coating based on the principle of quartz crystal microbalance (QCM). The new sensor can be used to detect the characteristic materials of a volatile liquid. A mechanical model of the new sensor was built, whose structure was a thin circle plate composing of polytef/quartz piezoelectric/polytef. The mechanical model had a diameter of 8 mm and a thickness of 170 μm. The vibration state of the model was simulated by software ANSYS after the physical parameters and the boundary condition of the new sensor were set. According to the results of experiments, we set up a frequency range from 9.995850 MHz to 9.997225 MHz, 17 kinds of frequencies and modes of vibration were obtained within this range. We found a special frequency f_{sp} of 9.996358 MHz. When the resonant frequency of the new sensor's mechanical model reached the special frequency, a special phenomenon occurred. In this case, the amplitude of the center point O on the mechanical model reached the maximum value. At the same time, the minimum absolute difference between the simulated frequency based on the ANSYS software and the experimental measured stable frequency was reached. The research showed that the design of the new polymer quartz piezoelectric crystal sensor perfectly conforms to the principle of QCM. A special frequency value f_{sp} was found and subsequently became one of the most important parameters in the new sensor design.

Keywords: quartz piezoelectric crystal sensor, quartz crystal microbalance, vibration analysis, special resonator frequency

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1. Introduction

A sensor made of quartz piezoelectric crystal is the main component of intelligent monitor equipment. Because of its simple structure, small volume, light quality, low consumption and long life, the piezoelectric sensor has been widely used in the area of food detecting, cosmetic and pharmaceutical industries for achieving real-time detection and early warning of product quality and ingredients during the production process; so the research of piezoelectric sensors holds great significance. Many scholars at home and abroad study quartz piezoelectric crystal sensors. For example, Tan and Huang[1] investigated the frequency shift of a quartz crystal microbalance (QCM) sensor induced by the mass effect and discuss the properties of the coated viscoelastic film. Vasile *et al.*[2] revealed that liquid can be detected without contacting by the out-of-plane vibration of a quartz crystal resonator. Hussain *et al.*[3] researched acidic and basic polymers for molecularly imprinted folic acid sensors. Lu *et al.*[4,5] performed an energy-trapping analysis for the bi-stepped mesa quartz crystal microbalance using the finite element method. Khasan[6] investigated the properties of displacement sensors based on polyaniline films. Chen *et al.*[7] researched the tunable characteristics of bending resonance frequency in magnetoelectric laminated composites. Gu[8] researched the structure and absorbability of a nanometer nonmetallic polymer thin film coating on quartz used in piezoelectric sensors. Based on the above studies, a new polymer quartz piezoelectric crystal sensor for detecting the characteristic materials of volatile liquid is designed, which takes a quartz piezoelectric crystal as the basal material and a nanometer nonmetallic polymer thin film as the surface coating and is based on the principle of QCM. We find a changed rule of the new sensor vibration frequency in the cleaning phase. The changed rule of the vibration frequency is important to the new polymer quartz piezoelectric crystal sensor design.

2. Design and test of the new sensor

A QCM measures the mass per unit area by measuring the change in frequency of a quartz crystal resonator. The resonance is disturbed by the addition or removal of mass deposited at the surface of the resonator. Considering the principal of the QCM, we know that the change in the oscillation frequency of the piezoelectric crystal is related with the mass deposited on it by the famous Sauerbrey equation

$$\Delta f = \frac{-\text{const.}\Delta m}{A} f_0^2 = N f_0^2, \tag{1}$$

where $N = -\text{const.}\Delta m/A$, f_0 is the resonant frequency of the quartz crystal, A is the piezoelectrically active crystal area, Δf

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is the frequency change, Δm is the mass deposited on the active area and const. is a bundle of constants describing the material properties of the piezoelectric crystal.



Fig. 1. Photograph of the new sensor.

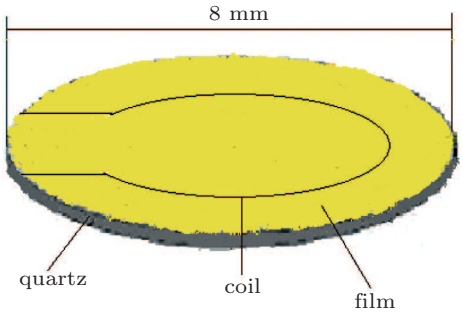


Fig. 2. (color online) Structural image of the new sensor.

We designed a new polymer quartz piezoelectric crystal sensor for detecting the characteristic materials of volatile liquid based on the principle of QCM. A photograph of the new sensor and an image of its construction are displayed in Figs. 1 and 2, respectively. For the new polymer quartz piezoelectric crystal sensor, an AT-cut quartz was used as the base and the polymer coating was deposited on the surface of the quartz with vacuum electron beam dispersion (EBD) coating technology.

For detecting the characteristic materials of a volatile liquid with the new polymer quartz piezoelectric crystal sensor, we carried out a series of experiments. GuoJiao 1573 (52 vol.%), one of the famous Maotai-flavor liquors, was selected as the standard sample of the volatile liquid in the test. Eight kinds of polymers were selected as the coating materials for the new sensor (Table 1). After analyzing the chemical compositions of the standard samples, we used the eight kinds of new sensors to compose a sensor array (Fig. 3). We could monitor the change in the frequency of the sensor array and capture the datum of change in frequency with a frequency counter. The working process of the new polymer quartz piezoelectric crystal sensor for detecting characteristic materials contains two phases: the cleaning phase and the adsorption phase. We concerned the change rule in the frequency of the new sensor in the cleaning phase. In this phase, the new sensor could be checked and corrected by the blowing of clean-dry air, since the increase in the new sensor frequency is caused by the decrease in the quality of the new sensor.

Table 1. Coating materials on different sensors of the sensor array.

Sensor number	1	2	3	4	5	6	7	8
Coating material	PVC	polyamide	polyethylene+AgCl	PTFE	AgCl	azithromycin	$CuCl_2$+PE	$CuCl_2$+AgCl+PE

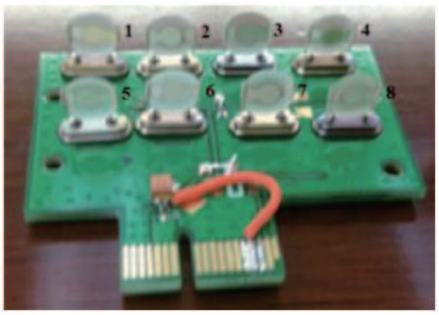


Fig. 3. (color online) Sensor array.

At room temperature, the frequencies of the eight sensors were monitored periodically with the use of a frequency counter and later recorded using new software. Figure 4 display the change in frequency of the new senor during the cleaning phase. We observed that with the extension of time, the frequency will increase at first and then stabilize. With the datum captured, we fitted them by using the least-square method and acquired a piecewise function, which is divided into a linear-nonlinear-linear style. The piecewise function can illustrate the change rule in the frequency of the new sensor during the cleaning phase.

The fitted curve in the Fig. 5 is the change rule in the frequency of the fourth sensor in the cleaning phase. The fourth sensor structure is composed of polytef/quartz piezoelectric/polytef. The piecewise function of the fitted curve can be expressed as

$$\begin{cases} y = 0.7691x + 9996330, & (0 < x \le 16\ \text{s}), \\ y = -0.00174x^2 + 0.25412x + 9996338.68, & (16\ \text{s} \le x \le 80\ \text{s}), \\ y = 9996348, & (x \ge 80\ \text{s}), \end{cases} \tag{2}$$

where x represents the time in units of seconds and y represents

the frequency in units of Hz. In the piecewise function, the change in frequency displays a linear trend in the range from 0 s to 16 s. The result is consistent with the principle of QCM perfectly, which can be explained by the Sauerbrey equation. In the range from 16 s to 80 s, the frequency displays a nonlinear trend with the increase of time. The result shows that, for the characteristic materials, the coating is close to clean by blowing clean-dry air as time goes on. After 80 s, the frequency reaches a steady state. The steady stage shows that the coating has reached a condition of cleaning and the quality of the new sensor does not change. We regard the stable frequency as the natural frequency of the fourth sensor. The stable frequency is one of the most important parameters for designing the new polymer quartz piezoelectric crystal sensors. The stable frequency of the fourth sensor has been obtained based on the above principles. It is $f_{st} = 9.996348$ MHz.

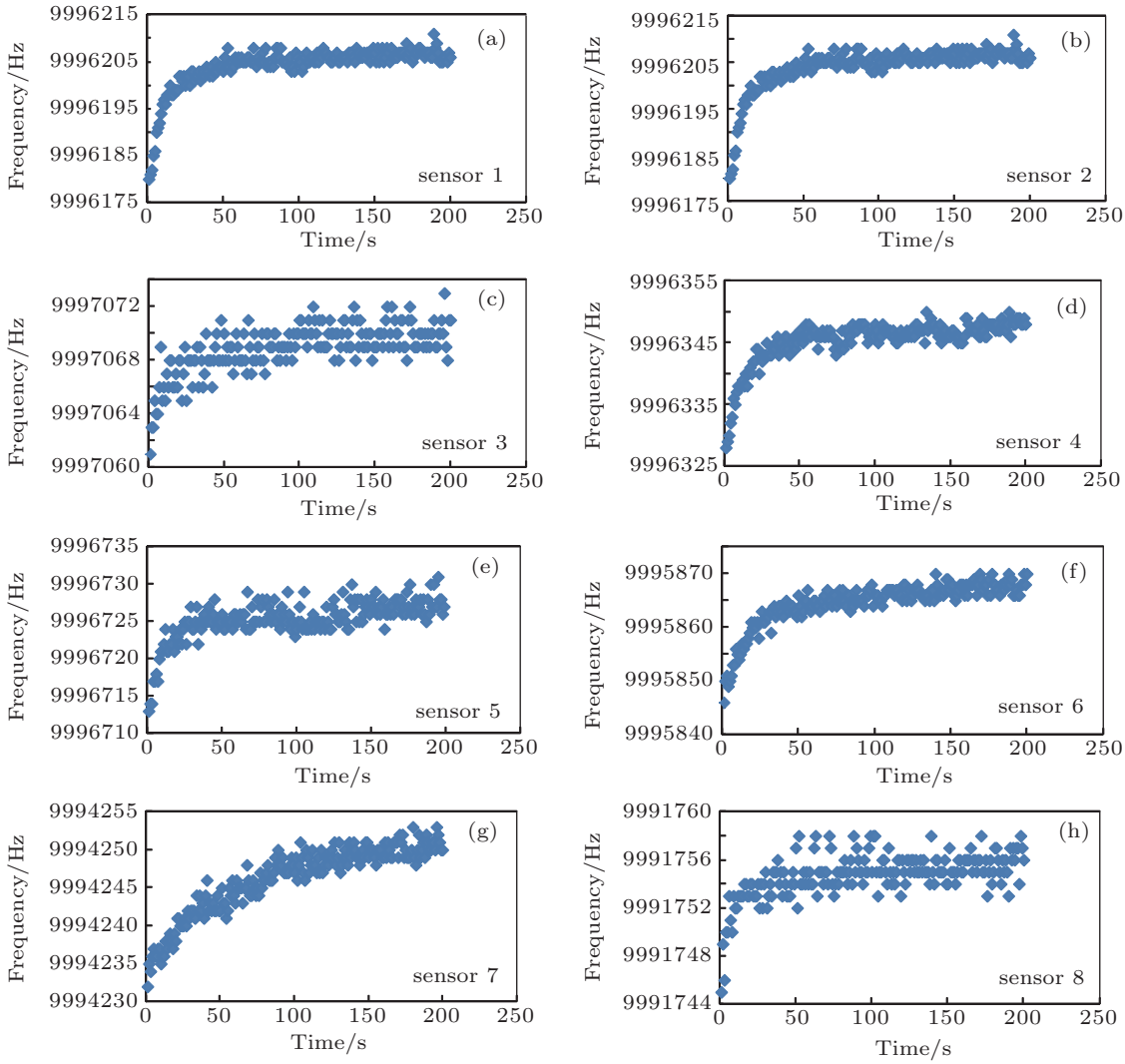


Fig. 4. (color online) Measured frequencies of sensors 1–8 shown respectively in panels (a)–(h).

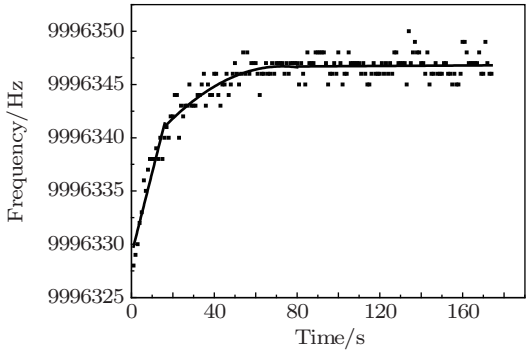


Fig. 5. Fitted curve of the fourth sensor.

We designed the fourth sensor with a structure of polytef/quartz piezoelectric/polytef based on the principle of QCM. As shown in Eq. (1), f_0 is the resonant frequency of the quartz crystal, so it is necessary to revise the Sauerbrey equation for the fourth sensor. We need to find a special resonator frequency f_{sp}, which belongs to the fourth sensor. Replacing f_0 with the special resonator frequency f_{sp} in the Sauerbrey equation, the revised equation can be obtained

$$\Delta f = \frac{-\text{const.}\Delta m}{A} f_{sp}^2 = N f_{sp}^2. \tag{3}$$

3. Vibration analysis of the model

As shown in Fig. 6, one mechanical model for the fourth sensor was built. The vibration of the model was simulated by software ANSYS after the physical parameter and the boundary condition were set. The geometric parameters of the mechanical model were confirmed. The radius of the new sensor r is 4 mm, the thickness of the quartz base h is 70 μm, and the thickness of polymer coating h_1 is 15 μm. Meanwhile, the boundary condition of the model is that both ends of the sensor are fixed at points A and B.

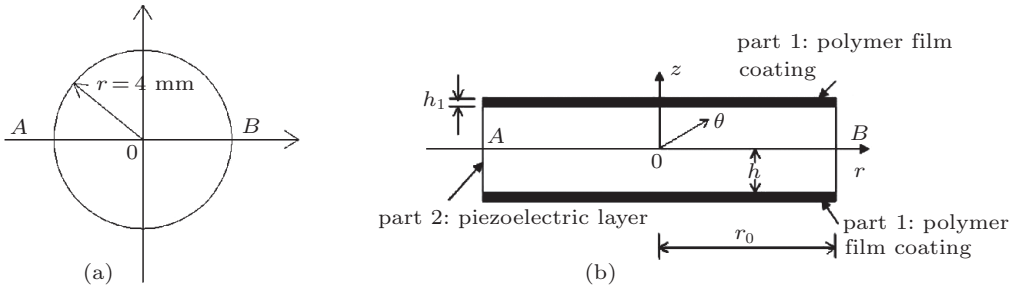


Fig. 6. Mechanical model of the fourth new polymer quartz piezoelectric crystal sensor: (a) vertical view and (b) front view.

The physical parameters of the AT-cut quartz piezoelectric crystal are listed as follows:

$$[\boldsymbol{C}] = \begin{bmatrix} 8.674 & -0.825 & 2.715 & -0.366 & 0 & 0 \\ -0.825 & 12.977 & -0.742 & 0.57 & 0 & 0 \\ 2.715 & -0.742 & 10.283 & 0.992 & 0 & 0 \\ -0.366 & 0.57 & 0.992 & 3.861 & 0 & 0 \\ 0 & 0 & 0 & 0 & 6.881 & 0.253 \\ 0 & 0 & 0 & 0 & 0.253 & 2.901 \end{bmatrix} \times 10^{10}\ \mathrm{N/m^2},$$

$$[\boldsymbol{e}] = \begin{Bmatrix} 0.171 & 0 & 0 \\ -0.171 & 0 & 0 \\ 0 & 0 & 0 \\ 0.0403 & 0 & 0 \\ 0 & -0.0403 & 0 \\ 0 & -0.171 & 0 \end{Bmatrix} \mathrm{N/V \cdot m}, \quad [\varepsilon] = \begin{bmatrix} 4.0 & 0 & 0 \\ 0 & 4.0 & 0 \\ 0 & 0 & 4.1 \end{bmatrix} \times 10^{-11}\ \mathrm{F/m}, \quad \rho = 7.6 \times 10^3\ \mathrm{Kg/m^3},$$

where $[\boldsymbol{C}]$ is the elastic matrix, $[\boldsymbol{e}]$ is the piezoelectric constant matrix, $[\varepsilon]$ is the permittivity matrix and ρ is the density of the quartz crystal. The physical parameters of the polytef are listed in Table 2.

Table 2. Physical parameters of the PTFE.

Material name	Elastic modulus/ 10^7 N·m^{-2}	Density/ 10^3 Kg·m^{-3}	Poissons ratio
PTFE	3	1.408	0.4

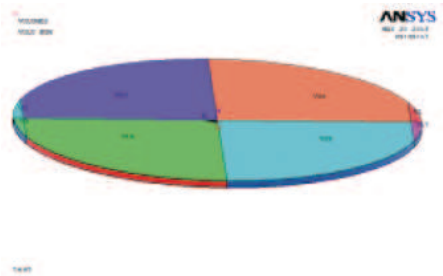


Fig. 7. The ANSYS model of the new sensor.

In order to obtain the vibration modal analysis of the new polymer quartz piezoelectric crystal sensor based on the ANSYS software, the fourth sensor's mechanical model was built, as shown in Fig. 7. The mechanical model was meshed through the ANSYS automatic mesh generation. The gridding of the model is shown in Fig. 8.

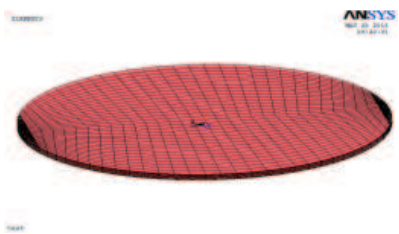


Fig. 8. The gridding of the model by the ANSYS.

We chose a range of vibration frequency from 9.995850 MHz to 9.997225 MHz in the ANSYS modal analysis based on a large number of experiments. In the frequency range, 17 different kinds of frequencies and modes of vibra-

tion were obtained. The first six orders of frequencies are the most influential parameters for the design of the new sensor. The obtained displacement nephograms of the model based on the ANSYS software are displayed in Figs. 9–14; the resonator frequencies f_r are shown in Table 3.

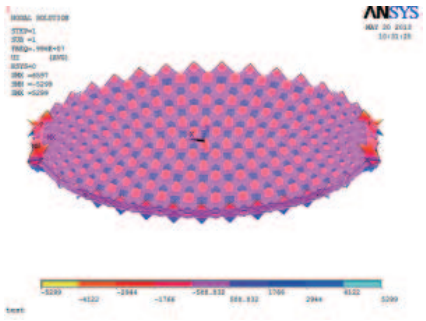


Fig. 9. Displacement nephograms of the first order.

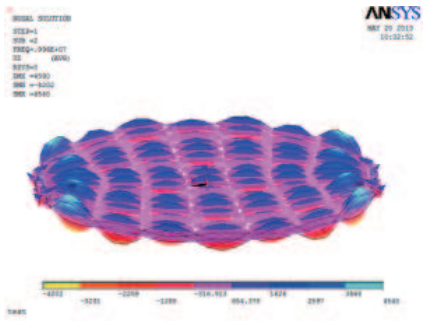


Fig. 10. Displacement nephograms of the second order.

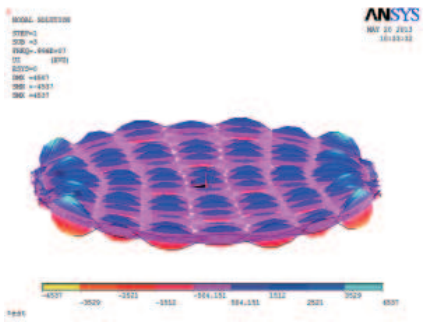


Fig. 11. Displacement nephograms of the third order.

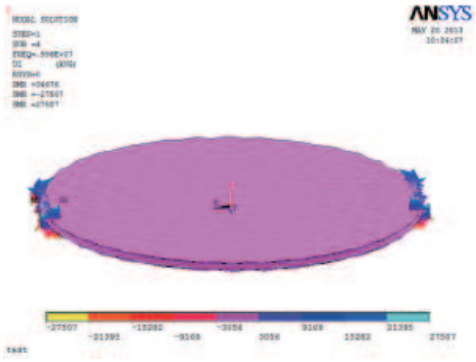


Fig. 12. Displacement nephograms of the fourth order.

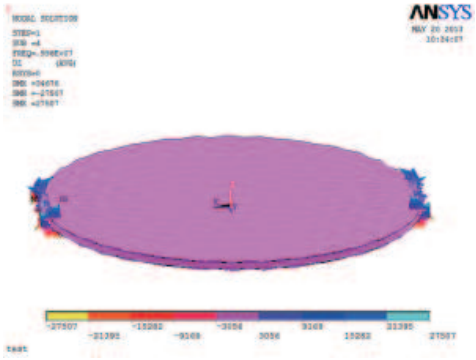


Fig. 13. Displacement nephograms of the fifth order.

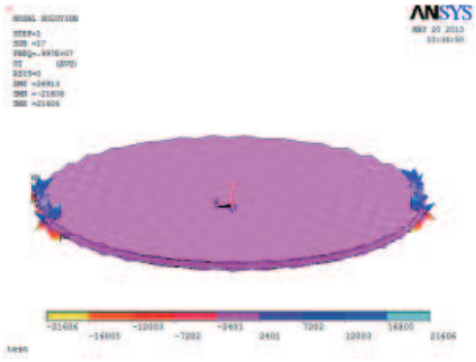


Fig. 14. Displacement nephograms of the sixth order.

Table 3. Absolute differences and absolute deviations of different orders.

Order	First	Second	Third	Fourth	Fifth	Sixth
Resonator frequency f_r/MHz	9.995890	9.995930	9.996358	9.996410	9.996490	9.997050
Absolute difference $f_r - f_{st}$/Hz	–457	–417	10	63	143	703
Absolute deviation $(f_r - f_{st})/f_{st} \times 100\%$	–0.69%%	–0.42%%	0.01%%	0.06%%	0.14%%	0.7%%

Table 4. Relative displacement at point O of the fourth sensor for different orders.

Order Node	First	Second	Third	Fourth	Fifth	Sixth
Relative displacement	1171.4	486.05	1690.6	1428	–1365.4	–1284.3
Resonator frequency/MHz	9.995890	9.995930	9.996358	9.996410	9.996490	9.997050

In Table 3, the absolute difference is the difference between the simulated frequency of different orders based on the ANSYS software and the experimental measured stable frequency of the fourth sensor. The absolute deviation is the ratio of the absolute difference to the experimental measured stable frequency. We find that both the absolute difference and the absolute deviation of the third order reach the minimum; the resonator frequency is 9.996358 MHz.

As shown in Table 4, the center point (O) of the thin circle plate on the model was extracted; its amplitudes were studied with the relative displacement. We find that the largest amplitude appeares at point O in the mechanical model in the third order and that the resonator frequency of the third order is 9.996358 MHz.

The special resonant frequency f_{sp} was 9.996358 MHz. When the resonant frequency of the fourth sensor reached the special resonant frequency, a special phenomenon occurred. In this case, the mechanical model's largest amplitude appeared and, simultaneously, the least absolute difference between the simulated frequency based on the ANSYS software and the experimental measured stable frequency was reached. So we can determine that the third order is the principal mode of the vibration in the mechanical model of the fourth sensor and that the special resonator frequency ($f_{sp} = 9.996358$ MHz) is the practical natural frequency of the fourth sensor.

4. Conclusion

The results presented here indicate that the new polymer quartz piezoelectric crystal sensor perfectly conforms to the principle of QCM. The new sensor can detect the characteristic materials of a volatile liquid. After the vibration modal analysis of the fourth sensor, whose structure was composed of polytef/quartz piezoelectric/polytef, based on the ANSYS software, we obtained the special resonator frequency of the mechanical model of the fourth sensor ($f_{sp} = 9.996358$ MHz). The absolute difference between the model's special resonator frequency and the new sensor's measured stable frequency is 10 Hz; the absolute deviation is only 0.01%%. The special resonator frequency value obtained by the ANSYS software can be seen as the practical natural frequency of the new sensor we designed. By replacing f_0 with the special resonator frequency value f_{sp} in the Sauerbrey equation, the revised equation can be obtained. The value is one of the most important parameters for designing the new polymer quartz piezoelectric crystal sensor used to detect characteristic materials of volatile liquid.

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