Study of a Crystal Sensor with Two Pairs of Electrodes

二つの電極を設けた水晶センサの検討

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1. Introduction

A differential method is applicable to the QCM system using two crystal sensors. However, the frequency temperature characteristics of two crystal sensors have a difference, respectively. There is a difference also in the stress of both crystals blank. It is high-cost. This QCM system has such a subject.

In order to solve these subjects, we proposed a new crystal sensor (twin sensor) by 2008 IEEE FCS^{1} . The characteristic of this sensor has two electrodes in same AT-cut crystal blank. And this is a sensor used as two-separation crystal resonator.

In this paper, the NAPiCOS system²⁾ is applied to this twin sensor, and the result of a detection experiment of CRP is shown. The NAPiCOS system is developed in the viewpoints of frequency stability, frequency temperature characteristics, noise, pulsation, etc., and is commercialized. CRP is one of the proteins currently studied in the medical field.

2. Experiment

The appearance of the twin sensor is shown in **Fig. 1**. The quality grade of the artificial crystal of an international standard is defined by inclusion density, an infrared absorption index, and dirty channel density. These apply the top quality grade article³⁾.

The design specifications of the sensor are two layers of a crystal blank: cut is AT, size is .⁹8.7, surface is polish, electrode material is chromium and gold, electrode size is 5.0mm (length) and 1.7mm (width), enclosure is glass-epoxy, and structure is the Langevin type.

Two electrodes, which a twin sensor has, are the same as that of MCF. The twin sensor applies the theory of this MCF. This theory is already generalized. For this reason, the details of the theory of the twin sensor are omitted. The elasticity combination between both electrodes of the twin sensor is eliminated. And the twin sensor has two-separation crystal resonators. This examination result is due to be reported to another opportunity.

The measurement accuracy of 1ng is secured in liquid. We set up such a temporary target. And in order to secure this frequency measurement accuracy, most design technique of crystal resonators adopt as the telecommunication system applied stress sensitivity, support stress, etc. Since a differential method is adopted, the electric merit of both the electrodes seen from the output terminal made it in agreement.

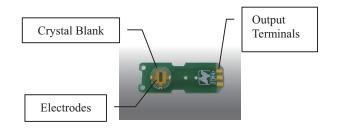


Fig. 1 Appearance of twin sensor

In order to realize an antigen-antibody reaction, CRP is applied to the twin sensor. The concentration: 100 ng/mL, amount of liquid for detection: 100 μ L, flow speed: 50 μ L/min, and buffer liquid: PBS 0.01Mol and pH 6.7 are applied.

The differential method is applied to the twin sensor. The twin sensor has an electrode by the side of a standard and a reaction. The standard side has the antibody blocked so that an antigen-antibody reaction may not arise. On the other hand, the antibody by the side of the sensor is not blocked. The jig for exclusive use is applied to formation of both this antibody and blocking.

Nominal frequency is 30MHz of fundamental vibration mode. The frequency change before and behind an antigen-antibody reaction is 11Hz (0.3

ppm). Mass change of these conditions is equivalent to about 1 ng by Sauerbrey equation. In measurement in liquid, 10,000 or less is the Q-value of the sensor⁴⁾. For this reason, in crystal oscillators apply to QCM, it is not easy to realize this 0.3ppm frequency measurement accuracy.

The twin sensor is dipped in liquid. The temperature stability of the oven applied to a sensor part is ± 0.01 degree C in preset temperature. The stirrer is not used. Change of the oscillation frequency by the stress sensitivity of the AT-cut crystal blank generates this reason.

The technology of the flow injection applied to LC/MS is used. The Anti-CRP monoclonal antibody is fixed at one of the two electrode of the twin sensor at the gold electrode. It block, in order not to react to one electrode of a twin sensor with the antigen of CRP. One electrode is not blocked. The antigen antibody reaction is made after blocking. And the oscillation frequency-time characteristics and Standard curve before and behind an antigen antibody reaction have been grasped. Furthermore, the check of CV value is performed.

3. Result and Discussion

Fig. 2 shows the oscillation frequency-time characteristics before and behind the antigen antibody reaction, which makes concentration of CRP 10 ng/mL. F1 is the frequency of a standard. This is the oscillation frequency by the side of the electrode, which does not react. F2 is the frequency of a sensor. This is the oscillation frequency by the side of the sensor, which reacts. F3 is these differences. F2 and F3 have overlapped in this figure. The antigen antibody reaction is made from F3. On the other hand, pulsation is not checked.

The finite difference method is applied. Viscous resistance by Kanazawa-Gordon equation, stress given to a crystal blank, other errors, change factors and etc is reduced or offset. And we think that F3 is an almost pure antigen antibody reaction ingredient.

Fig. 3 shows the analytical curve, which shows the result, measured in the range of 10 ng/mL to 10μ g/mL. The number of the used twin sensors is three respectively. For example, CV value of 100μ g/mL concentration is 1.28.

We checked the frequency amplitude at the time of an antigen antibody reaction, reaction time (joint time), the instability of frequency, and CV value change by the acquisition place of CRP. We believe that this results in the performance of CRP.

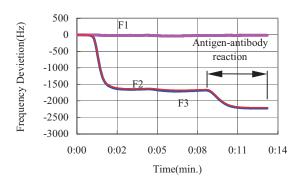


Fig. 2 the oscillation frequency-time characteristics before and behind

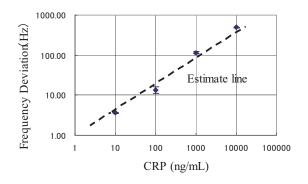


Fig.3 Standard-curve

4. Conclusion

In this paper, the twin sensor is applied to CRP. And the antigen antibody reaction is checked by experiment. The result checks that an antigen antibody reaction is possible also by the concentration of 10 ng/mL. Moreover, Standard curve and CV value is checked.

A future subject is the elucidation of the influence of the viscous resistance ingredient by a finite difference method, for example.

References

- 1. T. Muto, et., al., 2008 IEEE IFCS, pp.532-534
- 2. S. Wakamatsu, et., all, 2007IEEE IFCS, pp.16-19
- 3. IEC 60758
- 4. IEC 60444-1