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Mini Review

Quartz Crystal Microbalance (QCM) Sensing Materials in Biosensors Development

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Quartz crystal microbalance (QCM) biosensors are a type of analytical devices working on piezoelectric principle. The piezoelectric biosensors are different in their principle to the other types of biosensors, namely the voltametric and optical one because they measure a mass attached on their surface. It makes them readily for label free assays and construction of analytical devices where simplicity is expected and label free design is beneficial. Field assays and point of care tests are typical ways how a QCM based sensor will be applied in the praxis. On the other hand, the assays can exert significant specifications and be suitable for laboratory purposes. In this review, principle of piezoelectricity and QCM are explained and the recent applications of QCM in biosensors construction are described.

Keywords: affinity interactions; antibody; bioassay; immunoassay; immunosensor; label free assay; piezoelectricity; point-of-care test; quartz

1. INTRODUCTION

Biosensors became a relevant part of portable analytical devices suitable for performance outside laboratories, use for point-of-care diagnoses but also as a tool in the standard laboratories. Since the first electrochemical glucose biosensors, the various types of biosensors became a significant contribution to the total number of the available analytical instruments. The devices can serve for a wide number of analytes like biochemical markers [1-3], microorganisms [4,5], poisoning substances [6-8], drugs [9,10] or food components [11,12].

Biosensors are formed by a combination of a physical sensor and a biorecognition part (or biorecognition element in some sources) like an enzyme, an antibody, a receptor, or even whole cell, a sequence of DNA or RNA, and an aptamer etc. [13-15]. The sensor part can be typically optical or electrochemical but other types physical sensors like the electrochemiluminescence, gravimetric, non-liner optics exist [16-19]. This review is focused on the piezoelectric biosensors that use quartz crystal

as a material for the sensor construction. The idea of piezoelectric biosensors based on quartz crystal extensively developed in the last years and new devices with promising applicability were proposed. Principles and applications of the piezoelectric biosensors based on quartz crystals are surveyed here and pros and cons of the applications are discussed.

2. PIEZOELECTRICITY

Piezoelectric biosensors are a group of analytical devices working on the principle of piezoelectric effect respectively on the application of piezoelectric sensors. The common principle of piezoelectric biosensors is based on the piezoelectric phenomenon originally revealed by the Jacques and Pierre Currie. The piezoelectric phenomenon means that a piezoelectric material will be mechanically deformed when a voltage is imposed on his opposite sites or in a oppose manner, the material will generate voltage when it became mechanically stressed. The piezoelectric effect is common on the anisotropic crystals i.e. crystal with different physical properties in different directions. Materials like quartz, potassium sodium tartrate (Rochelle salt), tourmaline (boron silicate), zinc oxide, lead zirconate titanate, aluminum nitride, topaz (a mineral containing aluminum, fluorine and silicon), or organic polymers like polylactic acid, cellulose, polyamides, polyurea, and polyvinylidene fluoride exert significant piezoelectric properties [20-23].

In the analytical procedures, alternating voltage is lead to the piezoelectric material surface via attached electrodes and physical properties of the crystal are measured. Frequency of oscillation is the basic physical quantity to be determined. According to principle of the law of conservation of energy in the physics, energy kept in the oscillations should be constant hence attaching of a mass on the resonating piezoelectric material leads to the slowing of oscillations and removement of the mass from the piezoelectric material causes increasing of oscillations. Mathematical relation between oscillation frequency Δf and mass attached on the surface of a piezoelectric material Δm with density ρ, shear modulus μ and active surface square A expressed Sauerbrey in his equation as follow: $\Delta f = -2f_0^2 \Delta m / A(\rho\mu)^{1/2}$ [24-27]. The Sauerbrey equation is valid for an ambient environment not silencing the oscillation by a viscoelasticity. In the oppose case, the oscillation can be reduced proportionately to the density and viscosity of the environment. Resonances of the piezoelectric material in the liquids are a typical example. The relation between change of oscillations and density with viscosity defined Kanazawa and coworkers in their work [28-30]. The principle of the piezoelectric effect and measuring of mass bound to surface of a piezoelectric material is given as figure 1. The silencing of oscillations by viscosity of the environment may be a problem when a piezoelectric material is used for analytical purposes because. Matrix effect of samples can be for instance observed and dissipation of energy due to viscoelasticity of the environment should be considered when matrix effect influences measured signal.

The piezoelectric materials can work as microbalances and after modification of the active surface, the materials can serve for a construction of biosensors working on the principle of weighting analyte captured on their surface. The opportunity to construct an analytical device suitable for recording

the analyte without use of any specific reagents (label free assay) is the common advantage of the piezoelectric biosensors.

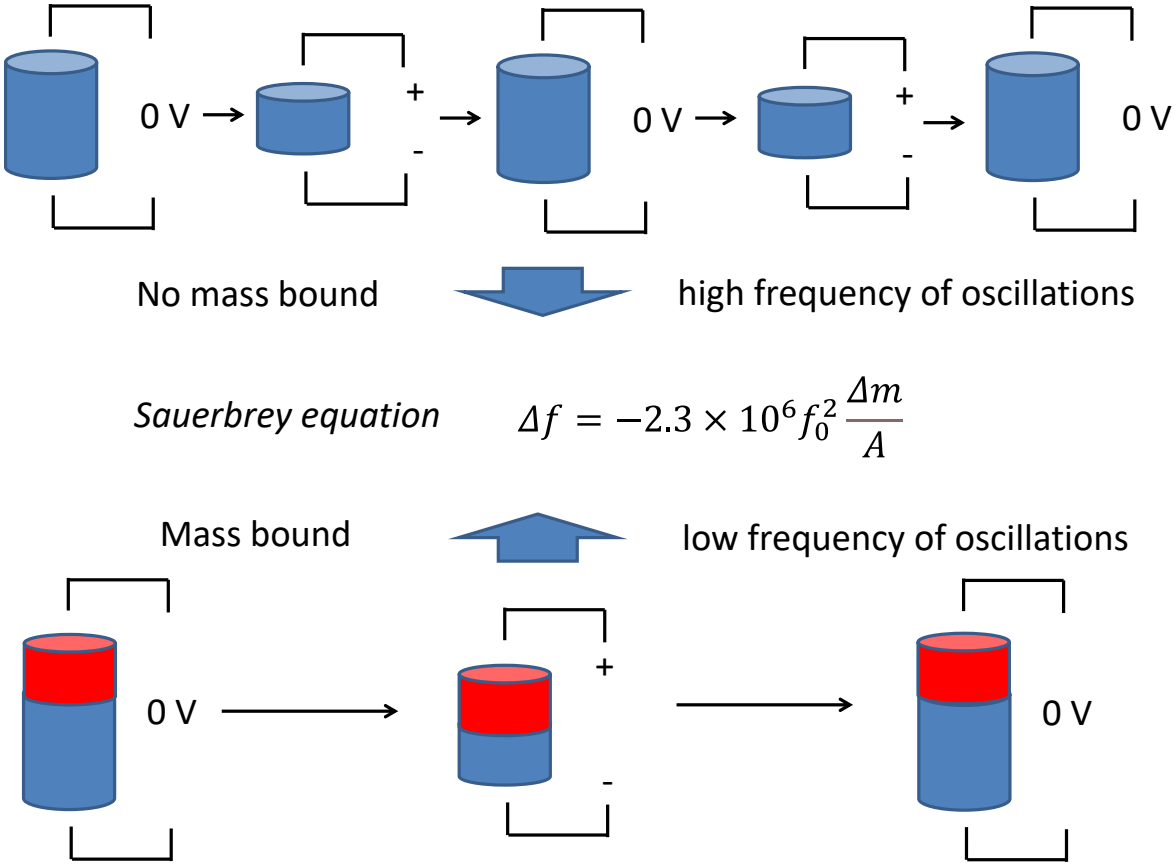

Figure 1. General principle of piezoelectric biosensors. A piezoelectric sensor is depicted as the blue cylinder while the bound mass as the red cylinder. In the Sauerbrey equation, Δf means change of equilibrium oscillation, f_0 is the original frequency of oscillation, Δm is mass bound to the piezoelectric sensor surface and A is the active surface where the oscillation occurs. This variant of equation is valid for quartz sensors, the constant 2.6×10^6 should be replaced by one submultiple where denominator contains root of material density fold by shear modulus of the used material.

3. QCM AS A MATERIAL FOR PIEZOELECTRIC BIOSENSORS

As mentioned in the text above, quartz is one of the piezoelectric materials and it finds a wide applicability in the analyses and electrotechnology as crystal oscillators / resonators and the use of quartz for this purpose is probably the most common. Various electronical devices for electrical metering, communication systems, network communicators, information technologies, clock generators, global position system detector devices etc. contains crystal oscillators made from quartz [31-36]. Quartz crystal oscillator is typically manufactured and sold under name quartz crystal microbalance (QCM) which is a thin cut of quartz crystal covered with metal electrodes on the opposite sites of the QCM. A common appearing of an QCM is depicted as figure 2.

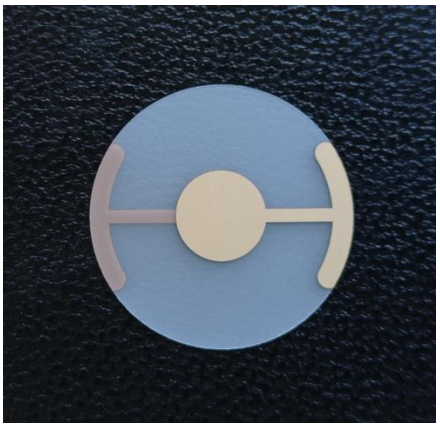

Figure 2. Appearance of a QCM sensor. A 10 MHz sensor based on 19 mm quartz circle with gold 7 mm wide electrodes on the opposite sites is depicted here.

In the most cases of QCM fabrication, the electrodes made on a quartz are composed from two metals. The underlayer serves for making a good mechanical and conductive contact between the electrode itself and quartz while the upper layer should be conductive as well as chemically stable to prevent oxidation and also suitable for connecting of the QCM into an electrical circuit (soldering or mechanical contact). Combination of gold in the upper layer and chromium as an underlayer exerts good stability and reproducibility and can be found in many applications. Other metals can be also used for a QCM manufacturing. Use of titanium as an underlayer and aluminum or silver as a material for the electrodes can be exampled [37-39]. The quartz platform for the QCMs is made by cutting of a quartz monocrystal rods. AT cut is the common type of cutting of monocrystal where angle from z axis is equal to 35.25º [40]. SC cut is another way how QCM sensors can be manufactured [41]. The SC cut is made under angle from z axis 34.11º resulting in similar properties to AT cut but the SC cut exerts better temperature and aging stability but the manufacturing process is more expensive. Other, but less common, cuts like BT, XY, GT and IT exist as well. Thickness of the cuts is in sub millimeter values when thinner cuts allow higher oscillation frequencies. Typical thickness for a 10 MHz QCM is 166 μm, the 5 MHz QCMs had typical thickness 330 μm and the 3 MHz have 500 μm [42]. The thickness drops exponentially with the increased frequency as thickness under 100 μm is necessary for frequencies around 20 MHz and 50 μm for frequencies around 30 MHz.

Because QCMs are mass-produced components of the electronics, they are quite cheap and eventually available in cost that allows its application elsewhere. The final price is mainly influenced by the cost for raw materials acquisition to the manufacturing process. The use for the chemical or biological analyses is not the main purpose the QCM are manufactured for but it can easily use them due to price and availability. The functionality of QCM in the construction of biosensors was proved in several studies and conclusions there were quite optimistic. The pioneer works devoted to the QCM biosensors were made in the end of 1980s and in the beginning of 1990s and various biorecognition parts like

antibodies and antigens or enzymes were proved to be suitable for the purpose of the biosensors manufacturing [43-50].

QCM devices with a basic oscillation frequency 10 MHz are used in the number of works but QCM with other basic frequencies are available as well. The thinner QCM have higher oscillation frequency but ore more elaborative for manufacturing and more fragile. Higher price is another disadvantage of the more sensitive QCM sensors with higher oscillation frequency. The final choice of the a QCM sensor for piezoelectric biosensor making is a compromise of several factors leading to the selection of a specific frequency. On the other hand, highly sensitive QCMs with oscillation frequency 3 MHz [42], 5 MHz [51], 6 MHz [52], 10 MHz [53-55], 16 MHz [56], 20 MHz [57], 27 MHz [58], 34 MHz [59], 40 MHz [60], 50 MHz [61,62], 55 MHz [63], 64 MHz [64], 125 MHz [65], 77 and 155 MHz [66], 170 MHz [67] and 180 MHz [68] can be exampled as possible platforms. The higher frequencies lead to improving of sensitivity and lowering of limit of detections. For instance, Pirincci and coworkers tested 5 MHz and 120 MHz QCMs as platforms for a biosensor suitable for aflatoxin detection [69]. While the 5 MHz QCM biosensors detected aflatoxin with limit of detection 1250 ppt, the 120 MHz biosensor had limit of detection significantly lower: 250 ppt. The theoretical sensitivity of QCM to a mass attached on its surface is given by the Sauerbrey equation. For instance, a 5 MHz QCM sensor exerts sensitivity 17.7 ng/cm^2/Hz [70]. It means that an electrode with an area 1 cm^2 will cause decrease of quartz oscillation 1 Hz when 17.7 ng of a mass is firmly attached to the electrode surface. Comparing to the aforementioned, 10 MHz QCM have theoretical sensitivity is approximately four times higher: 4.4 ng/cm^2/Hz and the 50 MHz QCM ten times higher: 0.176 ng/cm^2/Hz [71]. Practical sensitivity is of course lower as the attached mass will not act as an ideal firm film homogenously attached over the electrode, elasticity of the attached mass can occur etc. Temperature, humidity, vibrations and pressure on the QCM device can also affect an assay [72]. Size of electrodes also play an important role and larger electrodes are more sensitive according the Sauerbrey equation. On the other hand, manufacturing costs are higher as well. Survey of QCM basic specifications is given as table 1.

Table 1. Survey of QCM specifications

Specification	*Value or denomination*	*References*
material of crystal	pure quartz	[31-36]
materials of electrode	gold, titanium, aluminum, silver as upper layer, chromium as underlayer	[37-39]
typical thickness of the quartz cut	166 μm (for 10 MHz), 330 μm (for 5 MHz), 500 μm (for 3 MHz), 100 μm (frequencies around 20 MHz), 50 μm (frequencies around 30 MHz)	[42]
cuts of the crystal (angle from z axis when quartz monocrystal rod is cut)	AT cut (35.25º), SC cut (34.11º), other cuts can also occur (BT, XY, GT, IT)	[40,41]

typical oscillation frequencies	3 MHz, 5 MHz, 6 MHz, 10 MHz, 16 MHz, 27 MHz, 40 MHz, 50 MHz, 55 MHz, 64 MHz, 125 MHz, 77 and 155 MHz, 170 MHz	[42,51,52,56,58,60-67]
mass sensitivity	17.7 ng/cm^2/Hz (for 5 MHz QCM), 4.4 ng/cm^2/Hz (for 10 MHz QCM), 0.176 ng/cm^2/Hz (for 50 MHZ QCM)	[70,71]

4. APPLICATIONS OF QCM BIOSENSORS

In the recent years, there is a huge progression in the applications of QCM in biosensors construction and the piezoelectric platform was proven to be competitive to the standard types of biosensors dominantly based on the principle of voltammetry or optical methods. Survey of the aforementioned applications of QCM in biosensors development is written in table 2.

In a work by Gomez-Arribas, there was used a QCM platform for studying of interaction between phytohemagglutinin, a lectin from legume, and specific V_H antibody fragments [73]. In another study, saliva glucose detection based on QCM sensor that contained phenylboronic acid hydrogel membrane covered with polyethylene glycol membrane coated with carbon nanotubes [74]. The phenylboronic acid hydrogel membrane served for a specific oxidation of glucose while the upper polyethylene glycol membrane protected the QCM from adsorption of proteins. The biosensor exerted limit of detection 0.5 mg/l and limited interference by proteins like albumin, mucin and fibrinogen. An assay for the detection of immunoglobulins G class by a QCM biosensor with protein A was presented by Zhou and coworkers [65]. They used an AT cut of QCM with basic oscillation frequency 125 MHz and they were able to detect as low as 1 ng/ml of immunoglobulin G. Rodphukdeekul and coworkers prepared a QCM biosensor where gold electrodes were modified by self-assembled monolayer of 11-mercaptoundecanoic acid following by a monoclonal antibodies against *Porphyromonas gingivalis* and *Aggregatibacter actinomycetemcomitans* known as periodontal bacteria [75]. The authors were able to detect as low as 800 bacteria and the upper end of the biosensor´s dynamic range reached 2.32×10^6 cells.

An analytical device based on QCM can be also made using a molecularly imprinted polymer. Such conception chosen for instance Islam and coworkers for the detection of diterpenoid lactone andrographolide in an extract from *Andrographis paniculata* plant [76]. The manufactured biosensor had binding capacity for andrographolide 18 ng/cm^2. They also expressed limit of detection and limit of quantification for the assay as a mass per square centimeter of electrode and it was equal to 1.21 ng/cm^2 for limit of detection and 4.02 ng/ cm^2 for limit of quantification. In another paper, a QCM biosensor with molecularly imprinted polymer was developed for the detection of L-tryptophan [77]. The membrane was synthesized in situ and was made from polymethacrylic acid crosslinked with ethylene glycol dimethacrylate in which L-tryptophan was imprinted. The membrane exhibited adsorption capacity approximately 12 mg/g for L-tryptophan and the whole assay had limit of detection 0.73 ng/ml. Molecularly imprinted polymer for trichlorfon assay by a QCM biosensor was developed by Dayal and

coworkers [78]. They chose polyvinylidene difluoride as a material for membrane and performed in situ polymerization on a QCM in the presence of trichlorfon. The constructed biosensor had limit of detection 4.63 ppb and limit of quantification 15.8 ppb. Dynamic range of the assay went up to 250 ppb of trichlorfon.

Table 2. Survey of QCM biosensors

Description	*Analyte*	*Specifications*	*References*
QCM covered with phenylboronic acid hydrogel membrane (responsible for glucose detection) and polyethylene glycol membrane coated with carbon nanotubes (protection from unwanted adsorption of proteins)	glucose	limit of detection 0.5 mg/l and limited interference by proteins like albumin, mucin and fibrinogen	[74]
125 MHz AT cut of QCM covered with protein A	immunoglobulin G	limit of detection 1 ng/ml	[65]
QCM biosensor with immobilized monoclonal antibody	*Porphyromonas gingivalis* and *Aggregatibacter actinomycetemcomitans*	limit of detection 800 bacteria, dynamic range up to 2.32×10^6 cells	[75]
QCM with molecularly imprinted andrographolide	andrographolide	limit of detection 1.21 ng/cm^2, limit of quantification 4.02 ng/ cm^2, maximal binding capacity for andrographolide 18 ng/cm^2	[76]
QCM with molecularly imprinted L-tryptophan into membrane from polymethacrylic acid	L-tryptophan	adsorption capacity of membrane 12 mg/g, limit of detection 0.73 ng/ml	[77]
QCM with molecularly imprinted L-tryptophan into membrane from polyvinylidene difluoride	trichlorfon	limit of detection 4.63 ppb, limit of quantification 15.8 ppb	[78]

5. CONCLUSION

The QCM biosensors represent a competitive platform for analytical devices suitable for field tests of pollutants, point of care tests of biochemical and immunochemical markers and other analytical

applications. Though QCM biosensors are not so widespread like the other types of biosensors including the optical or voltametric one, they may become popular in the future due to simplicity of assay (label free affinity detection), suitability for miniaturization and possibility to make the assay readily to be performed by people without special training or education. Costs of the devices will also play a role. The mass production of QCMs for electrotechnical industry make them commercially available for other applications like the biosensors construction. Growing interest on QCM applications in various chemical and biological analyses can be expected in the future.

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