From conventional membrane electrodes to ion-sensitive field-effect transistors

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Abstract—*The theory concerning conventional membrane electrodes is often used as a starting point for the theoretical description of the operation of an ion-sensitive field-effect transistor. Although this results in a useful description of the device, a better view of the principles, and consequently the possibilities, of the new ion-sensitive device may result if the analysis is carried out in more detail, especially with respect to measuring concepts. This is presented in the paper. A careful comparison between the operation of a conventional membrane electrode and the i.s.f.e.t. leads to a definite classification of different types of i.s.f.e.t.s. These types are illustrated with corresponding measurements. In addition, the most appropriate application for each type is mentioned.*

1 Introduction

AT A TIME in which the development of various types of ion-sensitive field-effect transistors (i.s.f.e.t.s) is making such great progress (BERGVELD, 1972; MATSUO *et al.*, 1974; ZEMEL, 1975; MOSS *et al.*, 1975; BERGVELD *et al.*, 1976; KELLY, 1977), it is useful to consider this new device in a classification of more conventional ion selective membrane electrodes (RECHNITZ, 1975). The principal differences are especially of interest, and, consequently, also the principal possibilities of application. As the pH glass membrane electrode is the most common type of membrane electrode in use and quite well understood, theoretically, as well as practically, the concept of this electrode will serve as an illustrating example of a conventional membrane electrode in this paper (EISENMAN, 1967; BATES, 1973).

For the sake of clarity it is of great importance to keep in mind the following definitions for interfacial phenomena:

- (a) interfaces through which charge transfer is possible: such interfaces are called nonpolarisable, while the electrochemical reactions occurring at the interface may be reversible or irreversible
- (b) interfaces through which no charge transfer is possible: these interfaces are called polarisable.

In order to develop a useful sensor device, we focus our attention on polarisable or reversible non-polarisable interfaces. In the following Sections polarisable and reversible will, therefore, be used as excluding adjectives. The equilibrium state of a

reversible reaction can be fully described thermodynamically as long as a net current through the interface is some orders smaller than the equilibrium exchange current. In this case the currents of single-charge carriers in both directions exactly compensate each other. This is assumed to be the case in all reversible reactions mentioned in the following Sections.

2 The pH glass membrane electrode

In principle, the pH glass electrode consists of a glass membrane that separates two aqueous solutions, one with a constant ionic composition and the other having a variation in pH that has to be detected. Both aqueous solutions are contacted by electrodes: usually silver/silver-chloride (Ag/AgCl) or calomel ($\text{Hg}_2/\text{HcCl}_2$) in saturated KCl.

At both liquid/glass interfaces hydrogen ions will cross the boundary between the solution and the gel layers of the glass, separated by the relatively dry glass bulk (EISENMAN, 1967; BATES, 1973). The exchange of the hydrogen ions between the solution and glass phase makes it possible to describe the interfacial phenomena in general as reversible reactions. The result will be an ionic double layer at both glass interfaces with corresponding junction potentials. These potentials are partly due to a diffusion of ionic charges in the gel layer of the glass (diffusion potential) and partly due to ion exchange at the liquid/glass interface (boundary potential). The resulting electromotive force (e.m.f.), being the sum of the junction potentials of the cell, as described above, can be measured between the two reference electrodes. If the accuracy of a pH measurement is required to be 0.001 pH, this voltage must be measured with an accuracy of 0.058 mV, assuming an ideal sensitivity of 58 mV per pH unit. With a

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symmetrical scale on the measuring device of 6 units around pH7 the maximum voltage is ± 348 mV, so that the accuracy requirement of the measurement will be 0.01%. This means that the internal resistance of the measuring device has to be 10^4 times the internal resistance of the glass membrane, being fixed by the relatively dry glass bulk. It is essential for a measurement with an instrument having a resistive input (electrometer) that there be charge transport through the whole system and that all interfaces are reversible. This means that it is necessary that ionic transport occurs in the glass. It was found that in pH sensitive glass the sodium ions are the charge carriers (JOHNSON *et al.*, 1951). That this can occur without disturbing the system can be seen from the following calculations.

Considering the well known Corning 015 glass electrode, which has a composition of 72.2 mole percent SiO_2 , 21.4 mole percent Na_2O and 6.4 mole percent CaO , it can be calculated that a glass membrane of 0.1 cm^3 contains 10^{21} sodium atoms (this amount of sodium atoms follows from the density of the glass, which is 2.35 g/cm^3 and the molecular weight of the glass which is 60.2 g/mol).

A current through the glass membrane of 10^{-10} A transports about 10^{12} sodium ions per hour, which is thus a negligible amount with respect to the available ions.

It is essential to keep in mind that the glass membrane separates two aqueous solutions, both with a reference electrode to which the electrometer is connected and that in the system as a whole a small current flows continuously. For correct

measurements, all electrochemical reactions at the different interfaces have to be of a reversible type.

3 Glass membrane with a capacitive load

Instead of connecting the reference electrodes of the system with an ion-conducting glass membrane to an electrometer, as mentioned above, let us consider the situation where they are connected to a simple, but ideal, capacitor. This is shown in Fig. 1, in which ϕ_1 and ϕ_2 are the junction potentials at the glass-solution interfaces, as a function of pH_1 and pH_2 , respectively. It is assumed that the value of the pH is such that the surface charge of the glass is positive and further that pH_1 is higher than pH_2 . Positive charges are indicated with + and negative charges with -. The numbers of + and - charges located in the glass, the solution and the capacitor are arbitrary, but for the distribution throughout the system the amounts will be consequently increased or decreased depending on the modification of the system, described in the following Sections.

For the sake of simplicity, the potentials of the reference electrodes with respect to the solution are assumed to be equal to each other ($E_{\text{ref } 1} = E_{\text{ref } 2}$). The capacitor C will be charged to a voltage $\phi_2 - \phi_1$. If one of the pHs changes, the voltage $\phi_2 - \phi_1$ will also change, and, consequently, the charge at the capacitor will change. Usually, the pH of one solution is kept constant thus the corresponding junction potential, e.g. ϕ_1 , is also constant. This means that $\Delta\phi_2$ as a function of ΔpH_2 is responsible for charging or discharging the

Fig. 1 Representation of a glass membrane between two aqueous solutions with pH_1 and pH_2 ($\text{pH}_1 > \text{pH}_2$). The solutions are connected by means of two reference electrodes to a capacitor C

Fig. 2 M.O.S. transistor with input capacitor C_{ox} connected between two reference electrodes

capacitor. It is essential that changing the charge content of the capacitor as a function of $\Delta\phi_2$, charge will be transported to and from the capacitor plates. This implies that also in this situation, charge transport is necessary in the glass membrane and that reversible reactions occur at all interfaces.

If the dielectric of the capacitor also transports a current (leakage current) a continuous current flows, as in the case mentioned in Section 2. An exception with respect to the necessity of current flow in the whole system arises if the glass bulk is not an ionic conductor but an insulator. In this case, the potential difference $\Delta\phi_2$ charges the load capacitance C and the glass capacitance C_g . Only if $C_g \gg C$ will $\Delta\phi_2$ appear almost completely across C . This case is, however, very unrealistic, although a similar case will be dealt with in Section 6.

4 Glass membrane with an m.o.s.f.e.t. load

If, in the concept mentioned in Section 3, the capacitor is an m.o.s. capacitor, a new possibility of measurement is created, as can be seen in Fig. 2. Now the capacitor C_{ox} will be charged to the voltage $\phi_2 - \phi_1$, which again needs charge transport through the glass membrane during every change in ϕ_2 , due to a change in pH_2 . The equilibrium situation with a voltage drop $V_{C_{ox}}$ over the capacitor can be measured, however, without requiring continuous charge transport through the glass membrane. In this situation the battery V supplies the energy for a continuous measurement of the charge in the inversion layer of the m.o.s. transistor, which is, in fact, the charge q at one of the plates of the capacitor C_{ox} and is related to the voltage $\phi_2 - \phi_1$ simply by the equation

$$q = C_{ox}(\phi_2 - \phi_1)$$

The relationship between q and the current in the

m.o.s.t. circuit can also be simply calculated (SZE, 1969) and measured in principle by a resistor R and an amplifier A .

5 Glass membrane with an o.s.f.e.t. load

A special type of m.o.s. transistor is the so-called o.s.f.e.t. (BERGVELD *et al.*, 1976). This device operates essentially in the same way as an m.o.s. transistor, thus measuring a charge q in agreement with $q = C_{ox}V$. However, the gate plate does not consist of a metal, but is directly an aqueous solution in contact with the oxide. It is assumed at this point that there is no exchange nor adsorption of charge between the solution and the oxide, or, in other words, the contact is ideally polarisable. Thus the device operates as a pure potential sensitive device with a capacitive nature.

A second difference with the m.o.s.f.e.t. conception of Fig. 2 is that the charge at the 'liquid plate' of the capacitor is now a distributed charge in the form of a diffuse layer, the thickness of which depends on the concentration of the aqueous solution (Debye length). The system is represented in Fig. 3.

Referring to Section 3, the oxide capacitance C_{ox} will again be charged as a function of $\phi_2 - \phi_1$, which implies charge transfer through the glass membrane and the external circuit as soon as the value of $\phi_2 - \phi_1$ varies. The difference between the previously considered example and the example mentioned above is that in the latter case only one reference electrode of a reversible type is necessary. In the examples of Figs. 1 and 2 the absolute values of the potentials of the two reference electrodes did not influence the system response. In the case of Fig. 3, the absolute value of the reference electrode potential E_{ref2} gives rise to a certain bias and C_{ox} is charged to a voltage $\phi_2 - \phi_1 + E_{ref2}$, where only ϕ_2 is variable.

Fig. 3 Glass membrane with one reference electrode and an o.s.f.e.t.

Fig. 4 Glass membrane with conductive glass integrated with an o.s.f.e.t.

6 O.S.F.E.T. directly connected to the glass membrane

As in the example mentioned in Section 5, one reference electrode is left out, and the question arises as to whether the corresponding aqueous solution can also be omitted. The function of the solution between the o.s.f.e.t. and the glass membrane is only a conduction for charge to load the 'liquid plate' of the capacitor C_{ox} . It will be obvious that in principle the length of this conductor can be made zero. This means that the charge source and the charge sensor are then fully integrated at one side. This can be seen in Fig. 4.

The charge on C_{ox} is increased with respect to the case given in Fig. 3, because $\phi_2 + E_{ref2} > \phi_2 - \phi_1 + E_{ref2}$. In fact we are now confronted with a whole new measuring concept that requires special attention.

The glass membrane is only contacted at one side with an aqueous solution, resulting in one corresponding junction potential as function of the pH of this solution. The glass membrane and the silicon dioxide can be seen as one layer from which the bulk properties will, however, differ from place to place. At this point it is assumed, for the sake of simplicity, that no additional charge or dipole layers, such as oxide charge and interface states, are present in this composite layer.

From the viewpoint of measuring technique it should now be considered how a change in ϕ_2 as function of pH will give rise to a change in the charge in the inversion layer of the o.s.f.e.t. The capacitor to be charged in this case is fixed by the original oxide if the glass membrane is an ionic conductor as in the examples mentioned in the previous Sections. The glass membrane now operates directly as a plate of the capacitor C_{ox} , on which the charge is again a distributed one as in the case of the 'liquid plate'. Once again a change in ϕ_2 now results

in a charge transport in the external circuit of the system and in the glass membrane. The charge transport in the glass membrane can also be seen as a charge redistribution in the glass membrane.

Contrary to the examples mentioned in the previous Sections, the concept mentioned here will also operate if the bulk of the glass membrane is not an ionic conductor but a perfect insulator. The capacitor to be charged by $\Delta\phi_2$ is then fixed by the total layer of silicon dioxide and glass membrane. Now only a charge transport occurs through the external circuit of the system. As the capacitor to be charged is, in this case, smaller than the original C_{ox} , the charge at the capacitor will consequently be smaller, and the sensitivity of the measurement less. This is a well known phenomena of m.o.s.f.e.t.s: the thicker the insulation layer, the less sensitive the device is. In principle the device is, however, still sensitive for $\Delta\phi_2$, but it is remarkable that in this case no charge transport is necessary in the bulk of the glass membrane. This is the essential difference with all concepts mentioned in the previous Sections. The reason is that charge on the solution side of the glass is already directly on the capacitor. Only part of the counter charge has to move through the external circuit. The result is shown in Fig. 5. The total number of + signs again equals the total number of - signs as in the other Figures, due to the requirement of charge neutrality, but the distribution differs from that shown in the previous Figures.

Note that this case of decreased sensitivity is essentially the same as that of the exceptional case mentioned at the end of Section 3 (insulating bulk of glass membrane). For the case of an i.s.f.e.t. this situation is, however, very realistic, especially if the ion sensitive membrane is not a glass membrane but a polymer one.

Of course it will be obvious that if the silicon dioxide has properties similar to those of the glass membrane with respect to the ion sensitivity and selectivity, the glass membrane can also be omitted, so that the transistor structure then directly operates as an ion sensitive device. In fact this is the most satisfactory way to construct a new device, called i.s.f.e.t., which can become a reality as soon as the technological problems of making transistors with ion selective oxides that do not influence the transistor operation itself are overcome.

In practice, however, it appears that the thermal silicon dioxide, necessary for a well defined transistor operation, does not have adequate ion selective properties. Adding impurities like Al_2O_3 , Na_2O etc., as in the case of glass membranes, in order to create the required ion selectivity, may have a detrimental influence on the transistor properties of the device. Therefore, the most appropriate way of constructing an i.s.f.e.t. will probably be the concept mentioned earlier in this Section of using a separate ion-selective layer on top of the SiO_2 of the transistor. If this layer is conductive, its thickness does

Fig. 5 Glass membrane with insulating glass integrated with an o.s.f.e.t.

investigated (REVESZ, 1977; BERGVELD *et al.*, 1978b). In view of the previous Sections it can easily be seen under what conditions this manner of operation will be a real possibility.

In Section 6 it was shown that, although the oxide itself does not need to be an ionic conductor, an external circuit, connected by means of a reference electrode, is absolutely necessary for the transport of negative charge from the solution to the silicon. If, in this situation, the oxide should be an ionic conductor, a leakage current through the oxide may occur, which will, however, not directly influence the system electrically as long as the condition as mentioned in the introduction is fulfilled. However, as soon as this internal charge transport results in electrochemical reactions at the silicon surface, difficulties will arise. Up to now this surface was assumed to be ideally polarisable, but if electrochemical reactions occur at the silicon/silicon-dioxide interface it may play the role of a reversible or irreversible electrode. Irreversibility can, of course, not be tolerated, because it means that the device will continuously change its character. Observed drift phenomena and the limited lifetime of i.s.f.e.t.s, as mentioned in Section 6, may be the result of such irreversible reactions at the silicon surface.

If, however, reversible reactions occur at the silicon surface, a new measuring concept is created. The silicon surface itself may now play a direct role as an ion sensitive electrode. Redox reactions are a well known example of such reversible electrodes. With respect to the thermodynamical description of the system, as mentioned in Section 7, an additional

Fig. 6 Charge and potential distribution of the system silicon-oxide solution

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thermodynamic equilibrium will then be established at the silicon-oxide interface. This equilibrium will also control the charge density at the silicon surface even if no reference electrode is inserted in the solution. This means that this concept, which requires an ion conducting oxide and one or another reversible reaction at the silicon surface, will operate in a single-electrode mode. The development of such a device is more difficult than the device discussed in the previous Sections. There the oxide had to be an insulator, with selective ion sensitive properties (adsorption) at the solution/oxide interface while the silicon/silicon-dioxide interface had to be polarisable. However, the single electrode i.s.f.e.t. requires an oxide that has conducting properties, for ions or neutral species, while at the same time ions from the solution have to control selectively the process occurring at the semiconductor-oxide interface.

It will be obvious, however, that it may also be possible to omit the oxide completely, thus using a semiconductor which is directly selectively sensitive for a specific reaction with the electrolyte for example with respect to a redox equilibrium in the solution (BERGVELD *et al.*, 1978b; BROUWER, 1967). An interfacial oxide may, however, be necessary as a transition region to couple both requirements of ion selectivity and reversible field effect (ZEMEL, 1975).

Although possible mechanisms have already been discussed in the literature (REVESZ, 1977), convincing measurements have still not been obtained. Up to now the experiments have only been executed with silicon/silicon-dioxide, but it will be obvious that this will not necessarily be the most adequate choice. The problem is that the necessary choice of materials for the oxide as well as the semiconductor is difficult to ascertain.

Another possibility for single electrode operation is reported in the literature and based on modulation of the surface state density as a function of ionic concentration from the electrolyte (BERGVELD *et al.*, 1978a; DE ROOY *et al.*, 1978). In the previous Sections the existence of surface states was concealed for the sake of simplicity, but in practice the surface state phenomenon existing at the silicon-oxide interface of all m.o.s. devices is also a reason why i.s.f.e.t.s differ so greatly from conventional membrane electrodes. The existence of surface states in fact yields an additional parameter of the device, which can actually be modulated as known from literature (YEOW, 1975).

9 Conclusions and resulting applications

In the previous Sections four principal measuring concepts have been discussed for the detection of ionic activity by means of the use of an ion sensitive membrane, in the examples limited to a glass membrane, indirectly or directly coupled to an (m.)o.s.f.e.t. device

- glass membrane between two aqueous solutions; two reference electrodes; internal ionic conduction necessary
- glass membrane between two aqueous solutions; one reference electrode and one o.s.f.e.t.; internal ionic conduction necessary
- glass membrane directly on top of an o.s.f.e.t., one reference electrode; no internal conduction necessary; i.s.f.e.t. concept
- glass membrane directly on top of a semiconductor; due to internal conduction and reversible reaction at the semiconductor surface, no reference electrode necessary; i.s.f.e.t. single-electrode concept.

It depends on the application desired as to which concept will be most adequate for a specific case. It also depends on the materials that are available to fulfil specific requirements which of the four principal possibilities can be made optimal for the application. Most probably concept (c) will be a popular subject in the development of ion sensitive devices in the future. Also in this specific case one can distinguish various possibilities, as already discussed in the previous Sections and partly also mentioned in the literature (MATSUO, 1974; MOSS, 1975; KELLY, 1977).

If we restrict ourselves to the use of silicon, because the silicon technology is so well elaborated, a first layer of thermal oxide will always be necessary

Fig. 7 Change in the equivalent gate potential $\Delta V_{g \text{ eq.}}$ as function of pNa and pK for an i.s.f.e.t. with thermal silicon dioxide that was annealed with aluminium

to fulfil the requirements of stable field effect devices. In principle three possibilities can now be distinguished

- (a) activation of the surface of the bare thermal silicon-dioxide with impurities, in order to obtain ion selective properties of the surface
- (b) attachment of an additional insulating layer to the silicon dioxide, in order to create a top layer selectively sensitive for ions, due to specific adsorption, ion exchange etc.
- (c) attachment of a conducting top layer selectively sensitive for ions due to ion diffusion, exchange etc.

From each of these three concepts the next experiments show the actual possibilities. Illustrating the first case, aluminium was evaporated on top of the thermal silicon dioxide and annealed at 450°C in N₂ for 10 min. It appears that after etching away the aluminium the surface is extremely hydrophylic and achieves selective ion sensitivity. For a device with 50 nm silicon dioxide, pretreated as mentioned above, curves can be measured as shown in Fig. 7. Here the i.s.f.e.t. device was applied in a bridge configuration, where the electrolyte potential was controlled by zeroing the bridge. This potential is called the equivalent gate potential V_{geq} and plotted as function of the electrolyte concentration. As can be seen, the device has a selectivity for K⁺ ions over Na⁺ ions in the order of 10. The sensitivity for K⁺ ions is 47 mV/dec and for Na⁺ ions 36 mV/dec. Instead of activating the surface of thermal silicon dioxide by means of building-in impurities due to annealing, it is also possible to deposit a c.v.d. film

Fig. 8 Change in the equivalent gate potential ΔV_{geq} as function of pAg for an i.s.f.e.t. with a polymer top layer impregnated with AgI

of SiO₂, mixed with other oxides, e.g. Al₂O₃ and Na₂O. In this way, the composition of ion selective glasses, as in use for conventional glass electrodes, can be approached with a technology compatible with the usual silicon technologies, which is of great advantage for the manufacture of this type of i.s.f.e.t.

The second case was derived by using an impregnated polymer prepared from a suspension of 10% silicone-rubber monomer and 10% AgI, dissolved in *n*-hexane. A very thin layer was brought on top of the silicon dioxide by dipping the i.s.f.e.t. in the suspension, and afterwards polymerised for two hours at 50°C. The result appears to be a Ag⁺ sensitive electrode, as shown in Fig. 8.

Although the transconductance of the device decreases with increasing thickness of the attached membrane, the method of measuring with a balanced bridge configuration is masking this effect. The accuracy of the device becomes less the smaller the transconductance, and thus the additional insulating top layer is thicker. The relative small sensitivity for Ag⁺ ions is due to the nonoptimal chosen percentage of AgI. Other layers useful for this type of device are Si₃N₄, Al₂O₃ etc.

For the third possibility, i.s.f.e.t. devices were coated with a rather thick layer of carbon paraffin (1 : 1 w/w) containing 10% of Ruzicka Cl⁻ sensitive electroactive material (Radiometer product) (RUZICKA *et al.*, 1971; MESARIC *et al.*, 1973). Owing to the carbon, the layer is conductive, which means that the sensitivity of the i.s.f.e.t. device is independent of the thickness of the layer. Fig. 9 shows a result of such an i.s.f.e.t. from which the sensitivity for Cl⁻ ions can be seen. The sensitivity for Cl⁻ ions is equal to the theoretically expected 59 mV/dec, due to the fact that here we used a commercially

Fig. 9 Change in the equivalent gate potential ΔV_{geq} as function of pCl for an i.s.f.e.t. with a carbon-paraffin wax top layer impregnated with Ruzicka Cl⁻ active powder

developed electroactive material, contrary to the materials used in the two previous examples that had not yet been optimised and thus did not give the possible theoretical sensitivity.

The three aforementioned examples of possible i.s.f.e.t. configurations are only illustrations. In the future we can expect a large number of special i.s.f.e.t. devices, each with its own specific top layer on silicon oxide or with a special layer directly on top of a semiconductor resulting in single-electrode operation. The ion sensitivity and selectivity phenomena are governed by the interface with the aqueous solution in the same way as conventional membrane electrodes. However, the measuring concept is quite different from the usual method that uses two reference electrodes. Furthermore, an advantage is the potential for microminiaturisation, which may easily lead to a multi-ion-probe development. Also, the flat structure of an i.s.f.e.t. will be of great advantage for attaching additional membranes.

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