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Stable self-sustained potential oscillations across a membrane filter impregnated with triolein

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Abstract

When applying constant electric current, periodic self-sustained potential oscillations with high stability are observed across a membrane filter impregnated with triolein, placed between KCl and NaCl aqueous solutions. Stability, reproducibility and controllability of the oscillation are much improved by the application of constant electric current compared with those obtained without application of electric current. Relations among value of electric current, base potential and period of the oscillations are studied, and it is concluded that the oscillation period can be controlled by base electric potential. Effects of temperature on the oscillations are investigated, and it is shown that Arrhenius plots for both base conductance and conductance amplitude of each oscillation fitted well to an individual straight line and that values of activation energies are similar to those of bulk salt solutions. From all data obtained it is suggested that the oscillations occur as a result of rhythmic repetition of opening and closing of hole(s) in the membrane, which is due to breakdown and restoration of a part of the membrane.

Keywords: Self-sustained potential oscillation; Triolein; Membrane conductance; Aqueous salt solution; Temperature dependence

1. Introduction

Several types of self-sustained oscillations of electric potential have been reported, where aqueous solutions of salt were separated by a membrane filter impregnated with lipid having oleyl radicals—such as diolel phosphate (DOPH) [1–5], monoolein [6], triolein [7–10] or sorbitan monooleate [11]—and where constant external forces were applied, e.g. difference in kind or concentration of the salt, pressure and/or electric current.

Among them, self-sustained potential oscillations across a cellulose ester filter impregnated with DOPH have been extensively investigated. It

has been shown that long-period oscillations occurred between KCl aqueous solutions of different salt concentration separated by a DOPH filter, and that more stable short-period oscillations occurred when an electric current and/or pressure was applied to the high salt concentration side [3]. Oscillations have also been observed between salt solutions of equal salt concentration separated by a filter impregnated with DOPH when different kinds of salt were used at both sides of the membrane and/or an electric current was applied [5]. Theoretical explanations have been presented for this kind of oscillation in refs. [4,12].

In case of a filter impregnated with triolein,

which is a nonionic lipid in contrast with DOPH, it has been reported that self-sustained oscillations occurred between equimolar KCl and NaCl solution without any external stimuli [7–9]. Another experimental work has also been reported using triolein membrane formed in a single fine pore, where two types of periodic oscillation have been observed between KCl and NaCl solutions [10]. However, the stability of the periodic oscillation has not been so good [8,9]. And because effects of external stimuli to control the oscillations were not clear, there has been no way to control the oscillations. Therefore, in order to understand the phenomena of self-sustained oscillations in more detail, to clarify the oscillation mechanism and to make useful those phenomena to some practical applications, better reproducible, stable and controllable oscillations are essential. In this paper, the authors will show that periodic self-sustained oscillations with higher stability and reproducibility can be obtained by applying a constant electric current across a filter impregnated with triolein. The periods of this type of oscillations can be controlled by value of the applied current. The relationships between applied current, induced base potential and oscillation period were studied. The amplitudes of the oscillations will be discussed in terms of membrane conductance. Subsequently, the effects of temperature on the characteristics of the oscillations will be dealt with. Discussion about the mechanism of the oscillations is presented based on the experimental data obtained.

2. Experimental

Cellulose ester filter paper, 8.0 μm nominal pore size, 84% pore ratio and 150 μm thickness (Nihon Millipore Ltd., type SCWP) was immersed in triolein (Sigma Grade Approx. 99%). Triolein penetrated the filter easily, and the quantity of triolein absorbed in each filter was about 15 mg cm^{-1} . This value did not depend on the length of immersion time. Figure 1 is a schematic illustration of the experimental setup. A filter paper impregnated with triolein was placed between two acrylic cells by using two

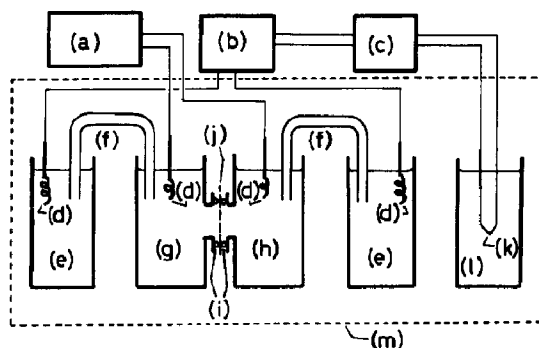


Fig. 1. Schematic diagram of the experimental apparatus. (a) Current source (Advantest TR6143), (b) electrometer (Advantest TR 8411), (c) pen recorder (Rika-Denki R-100) (d) Ag/AgCl electrode, (e) 3 M KCl aqueous solution, (f) KCl salt-bridge, (g) 0.5 M NaCl aqueous solution, (h) 0.5 M KCl aqueous solution, (i) silicon rubber gasket, (j) filter impregnated with triolein, (k) thermocouple, (l) distilled water for monitoring temperature, and (m) thermostatted air bath (Satake SC-T4-70C).

silicon rubber gaskets of 11.7 mm diameter. One cell was filled with 0.5 M KCl aqueous solutions and the other with 0.5 M NaCl aqueous solution. The volume of each cell was about 30 cm^3 . An electric current was applied from a constant current source with scanning function through two Ag/AgCl electrodes. The electric potential between the two aqueous solutions was monitored by an electrometer of high input impedance via a salt bridge combined and Ag/AgCl electrode, and recorded by a pen recorder. In the results shown below, the electric potential is represented as that of the KCl solution against the NaCl solution, namely, a positive value of the electric potential means that the electric potential of the KCl solution is higher than that of the NaCl solution, and a positive value of the electric current means that electric current is applied from the KCl solution to the NaCl solution.

Equipment depicted in the dotted cadre in Fig. 1 were placed in a thermostatted air bath. The temperature of the filter paper surrounded by salt solutions was monitored indirectly through another acrylic cell filled with distilled water placed in the thermostatted air bath. Experiments were begun at room temperature, $19 \pm 0.5^\circ\text{C}$. When effects of temperature on an oscilla-

tion were investigated, temperature was changed after the oscillation became stable.

3. Results and discussion

For two to seven days after a filter paper impregnated with triolein had been set between two acrylic cells filled with the salt solutions, the KCl side usually showed a negative potential of around -60 mV with respect to the NaCl side, and the membrane resistance kept a very high value of $1\text{--}2\text{ G}\Omega$. Under these conditions, oscillations of electric potential of small amplitude were observed occasionally without applying an electric current, although it rarely occurred. In addition, even when the electric current which induced an electric potential as much as 110 V was applied, neither oscillation nor change of membrane conductance occurred.

Then, after this initial period, the membrane potential suddenly reduced, and irregular upward (a direction for KCl side positive) impulses occurred. At the same time, the membrane resistance reduced to around $150\text{ M}\Omega$. Under this condition, when a constant electric current that induced a membrane potential of about 0.5 to 30 V was applied, stable and rhythmic oscillations of the electric potential occurred immediately or some time after [13]. Values of the electric current that induced the oscillations were in the range of 20 nA to $20\text{ }\mu\text{A}$.

3.1 General characteristics of oscillations

Typical oscillation curves are shown in Figs. 2, 3 and 4. Period and amplitude of each oscillation were stable for at least several hours, and in some cases for more than 10 days. Oscillations became such stable ones by application of electric current. The situation that the application of electric current played an important role to stabilize oscillations is similar to the case of DOPH [2–5]. Figures 2, 3 and 4 show downward (i.e., in the direction that reduces the absolute value of the electrical potential) pulse type, bistable type and sawtooth type of oscillation curve, respectively. These three types are typical curves of stable

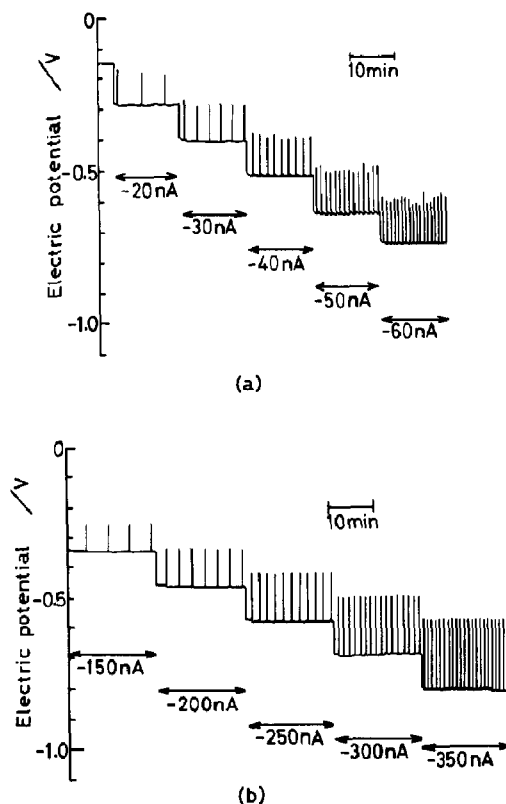


Fig. 2. Curves of stable oscillation of the electric potential, under application of an electric current of which the values are indicated in the figure. These are characteristic oscillation curves of the downward pulse type. (a) An oscillation curve taken 2 days after the oscillation began. (b) An oscillation curve taken 14 days after the oscillation began. Negative values of the potential and the current means that the current was applied from the NaCl side.

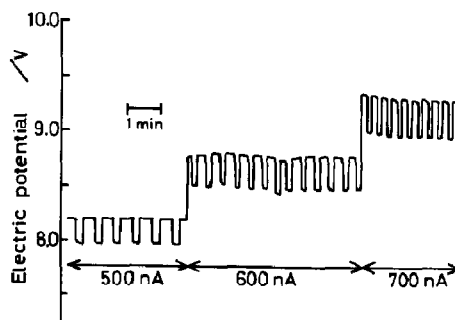


Fig. 3. A typical oscillation curve of the bistable type. Values of the current applied are indicated in the figure.

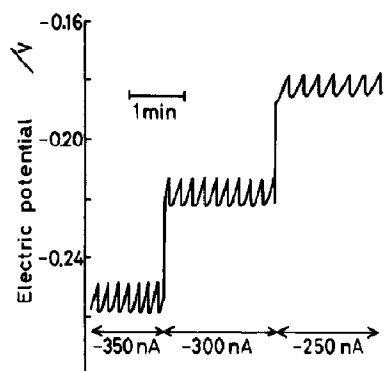


Fig. 4. A typical oscillation curve of the sawtooth type. Values of the current applied are indicated in the figure.

oscillations, and the downward pulse was generally the most stable type. Upward pulse also sometimes occurred and continued for a few hours, but base potential and amplitude of this type was generally less stable than of the others, and was not sustained long.

The mode of oscillation (or in other words the shape of an oscillation curve, the range of period and the electric potential level where the oscillation continued with high stability), differed from one sample to another, and it sometimes changed in one sample. It was difficult to control the mode of an oscillation, and it generally varied from one experiment to another. The direction of current which induced an oscillation also varied among samples.

As can be seen from Figs. 2(a), 2(b), 3 and 4, the oscillation period seems to be controlled by

the value of the current applied. However, when the relationships among oscillation period, applied electric current and base electric potential were investigated for many samples, it was revealed that oscillation period depended rather upon base electric potential (higher side of absolute value of the oscillating electric potential) than upon electric current. As an example, let us compare Fig. 2(b) with Fig. 2(a). The oscillation in Fig. 2(b) was obtained 12 days after that in Fig. 2(a) from the same sample. Since base membrane conductance in Fig. 2(b) became considerably larger than that in Fig. 2(a), the values of the applied electric current necessary to induce oscillation in Fig. 2(b) were much larger than those in Fig. 2(a). However, oscillation periods under similar level of base potential were similar in both cases as shown in Figs. 2(a) and 2(b). Similar results were obtained in some samples where temporary drops of base potential occurred during an oscillation [13]. In such cases, while base potential dropped and kept a certain lower level, period of oscillation kept a lower value which corresponded to the base potential for a smaller value of applied current. Figure 5 shows an example of plots of oscillation period versus base electric potential. In Fig. 5, 165 data from the same sample as that of Fig. 2 are plotted, which were obtained during 31 days of experiment. Plots fitted well to a straight line on a logarithmic scale. This means that the oscillation period was a power function of the base potential for all the data obtained during a long time of duration of

Table 1

Parameters of the oscillations in Figs. 2–4. These parameters are obtained by using not only data shown in each of the figures but also data at some other values of the current applied omitted from these figures

Oscillation	a	G (μS)	ΔG (nS)	S_h (μm^2)
Fig. 2(a) } Fig. 2(b) }	-1.99 *	$\left\{ \begin{array}{l} 0.079 \pm 0.005 \\ 0.433 \pm 0.003 \end{array} \right.$	$\left\{ \begin{array}{l} 25 \pm 7 \\ 172 \pm 11 \end{array} \right.$	$\left\{ \begin{array}{l} 0.79 \pm 0.23 \\ 5.51 \pm 0.34 \end{array} \right.$
Fig. 3	-1.92	0.0816 ± 0.0025	7.02 ± 1.02	0.179 ± 0.026
Fig. 4	-1.43	1.36 ± 0.01	58.3 ± 2.3	1.87 ± 0.07

Legend: a —Slope of the plots of oscillation period versus base electric potential on a logarithmic scale; G —Base membrane conductance; ΔG —conductance amplitude; and S_h —area of a hole estimated by assuming that the oscillation occurred as a result of opening and closing of the hole through the membrane with thickness of 150 μm .

* This value was calculated by using 165 data during 31 days.

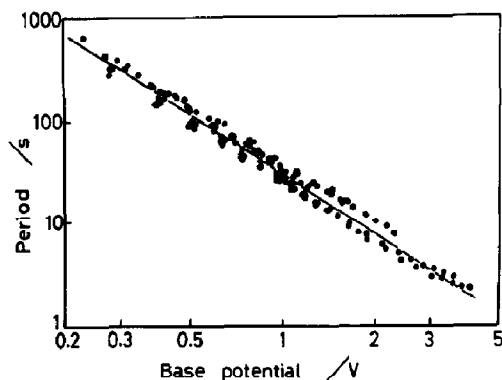
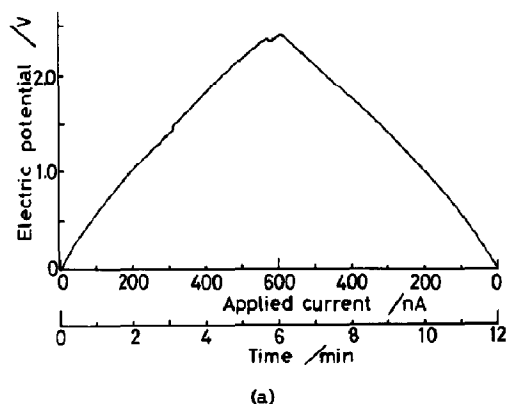


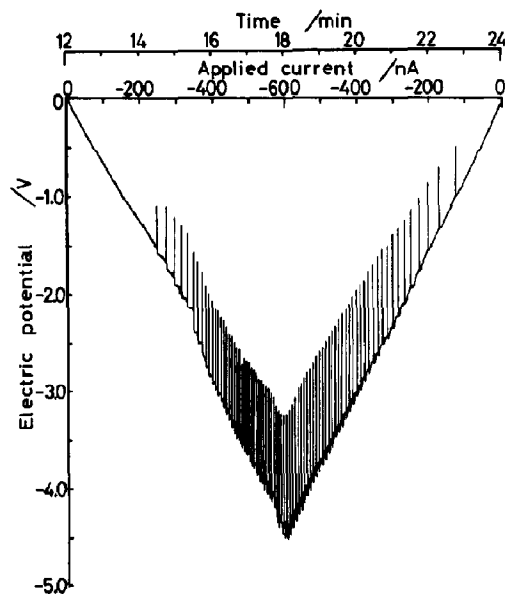
Fig. 5. Plots of oscillation period versus base electric potential for all the data obtained while the oscillation continued for 31 days. Sample is the same as that of Fig. 2.

the oscillation despite of an increase in base conductance. In Fig. 5, a slight shift of the line to longer period side is observed. However, this shift occurred within two days after this oscillation began, and all the other data obtained after these

two days fitted well to a single line. In case of Fig. 5, the slope of the line was -1.99 , which is listed in Table 1 together with those for the oscillations shown in Figs. 3 and 4. The slope was different from one mode of oscillation to another and it was between -1.3 and -3.5 for 90% of 54 modes in 25 samples investigated. The maximum and the minimum values of the slope were -4.6 and -0.9 , respectively. Such values of slope were generally larger than that of the period–current relationship obtained by Iiyama et al. [5], who reported the value of -1 for a filter impregnated with DOPH. In the present case, the plots of oscillation period versus applied current also fitted to a straight line on a logarithmic scale, when only data before changes in membrane conductance above mentioned occurred were used. Generally, the slope of each period vs. current relationship was slightly smaller than that of period vs. potential relationship because of nonlinearity of current vs. potential relationship of the membrane (see Fig. 6).



(a)



(b)

Fig. 6. Electric potential–current relationships for the same sample as that of Fig. 2, in case that the electric current applied was scanned at a constant speed. (a) Electric current was applied from the KCl side. (b) Electric current was applied from the NaCl side immediately after experiment (a).

Usually, after an oscillation started, base membrane conductance, G , was between 10^{-2} and $1 \mu\text{S}$, and the amplitude of an oscillation expressed in terms of conductance amplitude, ΔG , was between 10^{-3} and $10^{-1} \mu\text{S}$. The G and ΔG of the oscillations shown in Figs. 2–4 are listed in Table 1. In some cases, G and ΔG somewhat depended on applied current. So the values in Table 1 are averaged for several values of applied current. Both G and ΔG varied within three orders of magnitude from one oscillation to another as mentioned above. However, no specific relationship between either of them and shape of oscillation curve, period of oscillation nor direction of electric current to induce the oscillation was observed.

Figure 6 shows an oscillation curve of the same sample as Fig. 2 where electric current was continuously increased and decreased. First, an electric current from the positive direction was increased continuously and then decreased to zero (Fig. 6(a)), and after that a current from the negative direction was applied (Fig. 6(b)). As can be seen from the figure, the oscillation did not appear at all when the positive current was applied, and oscillation reappeared when switching back to a negative current. The direction of applied electric current which could induce stable oscillations varied from one sample to another, and in some samples both directions of current induced stable oscillations. However, even in such cases, the oscillations did not occur alternately by changing the direction of the current, and the mode of oscillation induced by one direction of the current was generally different from the other induced by the opposite direction of the current. That is to say, in such samples, one mode of oscillation was induced by one direction of current, and when the direction of current was reversed, the oscillation disappeared, and after that oscillation had completely disappeared by lapse of time or some other causes even when electric current of the former direction was applied, then another mode of oscillation occurred by applying electric current of opposite polarity. In our experiment, among 54 modes in 25 samples, 32 modes in 18 samples were induced by electric currents of positive direction and 22 modes in 15 samples

were induced by those of negative direction. Consequently, eight samples among them showed oscillations for both directions of applied current.

3.2 Mechanism of the oscillation

The generation of stable oscillations of the electric potential under application of an electric current can be explained by periodic change of conductance of the membrane. When a constant electric current is applied across a membrane, an electric charge accumulates on the surface of the membrane and penetrates into the triolein present in the pores. If some parts of triolein in some pores are thinner than in all other parts, the electric current concentrates at the thinner parts and penetration of electric charge should be promoted there. Then the parts become gradually thinner, and as a consequence, the electric field across the thinner parts becomes much higher than across all the other parts of the membrane. When the electric field across the weakest part of the membrane exceeds some critical value, dielectric breakdown occurs at that part, and membrane conductance becomes higher abruptly. After the breakdown has occurred, the electric field across the membrane diminishes. In addition, since triolein is fluid, the broken pore can easily be filled with triolein near the broken part within a short period of time upon which the membrane conductance recovers to previous level. However, once dielectric breakdown has occurred at the weakest part of the membrane, triolein at that part becomes thinner and weaker than at all other parts of the membrane, even when conductance at that part recovers to the previous level. Therefore, if a constant electric current is applied continuously, repetition of the breakdown and the restoration occurs at the same part of the membrane.

By assuming that the oscillations occur as a result of opening and closing of a hole through a membrane, like a channel the area of the hole, S_h , can be estimated from the value of conductance amplitude. Values of S_h were calculated to be in the range of 0.1 and $3.3 \mu\text{m}^2$ in all cases of the oscillations observed. For the cases of the oscillations of Figs. 2–4, the values of S_h are listed in Table 1. However, it must be noted that

these values are estimated by assuming that a cylindrical hole of constant diameter is opened all through the filter of 150 μm thickness, and this assumption is ideal. Because the Millipore filters used here are made of entangled cellulose ester fibers, the shapes of the pores should be more complicated. Besides, according to the mechanism of oscillation discussed above, the triolein layer at those parts that contribute to the oscillation is probably thinner than at all other parts of the membrane. Then, by supposing that the thickness of the triolein layer that contributes to the oscillations is, for instance, 1/10th of the thickness of the filter, S_h becomes 1/10th of that mentioned previously. It is important that the estimated area of a hole contributing to the oscillations always be much smaller than the nominal pore size of the filter.

When the electric current is increased, the period of oscillation becomes shorter as shown in Figs. 2–4 and 6. This is easily explained by the mechanism proposed here, since the accumulation and the penetration of electric charge become faster as electric current increases. Without electric current, it becomes very difficult to obtain stable and reproducible oscillations of electric potential. Application of an electric current is necessary of stable and reproducible oscillations of electric potential to occur. The high electric resistance and the fluidity of triolein may also be important factors for the oscillations.

The mechanism of oscillation of electric potential proposed here is somewhat similar to those proposed by Yoshikawa et al. [11], and Toko et al. [4], and it is different from that proposed by Kawakubo [12]. However, because Yoshikawa et al. did not apply electric current, membrane potential was much lower than ours. In the model of Toko et al., the net ionic charge of the lipid (DOPH in their case) and the difference in ionic concentration of the two solutions at both sides of the membrane are important factors for the generation of an oscillation of electric potential, and both of these conditions are different from our case. Therefore the mechanism of our case may be different from both of those proposed previously. However, there is a possibility that, as Toko et al. pointed out for the DOPH mem-

brane, phase transitions of triolein may contribute to the breakdown and the restoration of a part of the membrane also in the present case. Phase transitions may also contribute to the sudden decrease of membrane resistance before the oscillations occurred.

The temporary drop of base potential shown in ref. [13] and the permanent increase of base membrane conductance with time as shown in Fig. 2 can be attributed to an opening (and a closing in case of a temporary drop) of holes at other parts of the membrane which behave as a bypass of electric current. When this opening and closing of holes also happens to be periodically, it appears as a mixture of two oscillations. If the holes are no longer restored, a permanent increase of base conductance is the result, as shown in Fig. 2. When such increases of base conductance occur, the electric current to keep the electric potential at the same level as before must be increased. If the electric potential is kept at the same level by increasing the value of the electric current applied, the electric current which flows in the part contributing to the oscillation remains constant even when the net membrane conductance changes. This is the reason why the period of oscillation depends on base potential rather than on value of applied electric current.

As for the oscillations shown in Figs. 2–4 and 6, it is reasonable from the shapes of each oscillation curve to suppose that the part of the membrane which contributes to each of the oscillations is a single pore. The temporary drop of base potential could be explained as opening and closing of another hole and consequently as a temporal generation of a second path of electric current occurring independently from the main oscillation. However, sometimes, rather complicated oscillation curves were observed as shown in Fig. 7. The lifetime of such oscillations was generally not long and these oscillations soon changed to more simple ones. In such case, there is a possibility that two or more pores contribute to the oscillation cooperatively by entrainment which is known as a general phenomenon in nonlinear oscillation systems.

The reason why an oscillation disappeared when the direction of electric current was in-

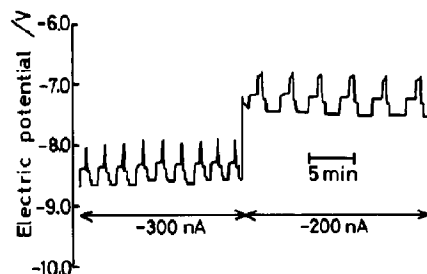


Fig. 7. An example of an oscillation curve which is complicated, but periodic and of single period.

verted as in Fig. 6, and the reason why the direction of electric current to induce an oscillation varied from one sample to another, are still unknown.

Next, in the following section, the authors will present and discuss experimental results of temperature dependence of the oscillations, which are completely compatible with the mechanism of oscillation presented here.

3.3 Effects of temperature change

Figure 8 shows a typical change of oscillation curve when temperature was gradually changed without changing the value of the current applied. As temperature rose, base potential became lower. At the same time, the period of oscillation became longer. Oscillations usually disappeared below 3 to -3°C , which probably relates to the melting point of triolein. In high temperature side, they disappeared at a temperature around 40°C , which varied in a rather wide range, 30 to 45°C , from one oscillation to another. This variation may be a result of not only temperature but also the lifetime of each oscillation.

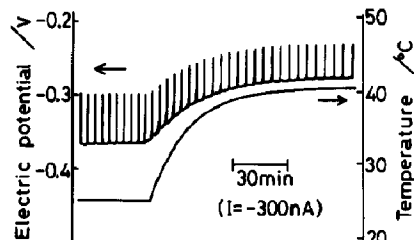


Fig. 8. A typical type of change of oscillation curve when temperature was gradually changed without changing the current applied.

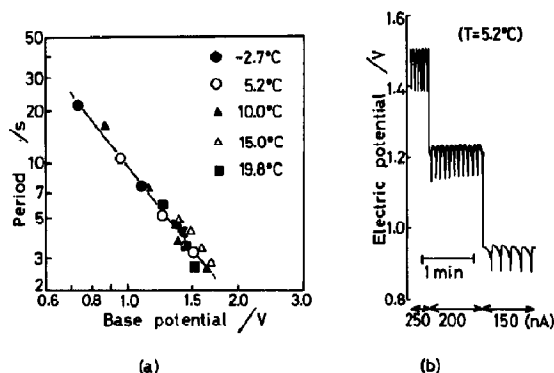


Fig. 9. (a) Typical plots of oscillation period versus base electric potential for various temperature conditions. No temperature dependence was observed for this oscillation. (b) Oscillation curve.

tion. It seems that a change of temperature, in other words "thermal stress", decreases the lifetime of an oscillation.

3.3.1 Period-potential relationships

Two types of temperature effect on period-potential relationships existed.

One type is shown in Fig. 9 together with its oscillation curve. In this type, the period-potential relationship had no temperature dependence. As temperature increased, base potential decreased and the period of oscillation increased under a certain fixed value of the current applied. In this case, a change of temperature did not affect the period-potential relationships. Change of temperature only altered base potential and, as a consequence, oscillation period. The oscillation shown in Fig. 8 is also of this type.

The other type is shown in Fig. 10, where the line of the period-potential relationship generally shifted to the lower side upon temperature increase. This means that as temperature rose, the period of oscillation at some specific level of base potential decreased. In this case, the oscillation itself was affected by temperature. The slope of the period-potential relationship showed a tendency to increase upon temperature increase. The temperature dependence of the dynamic viscosity of triolein seems to contribute to this temperature dependence. That is to say, when the temperature rises, the viscosity of triolein decreases, and then the breakdown and the restora-

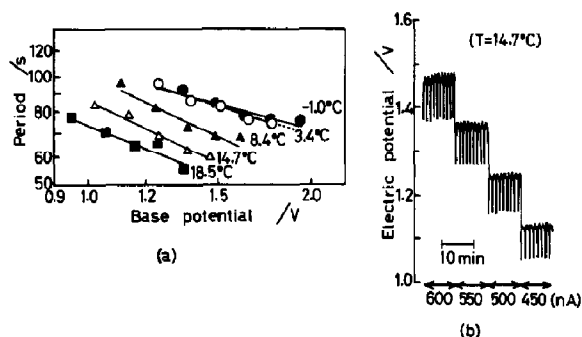


Fig. 10. (a) Typical plots of oscillation period versus base electric potential for various temperature conditions. The line shifted to the shorter-period side upon temperature increase. (b) Oscillation curve.

tion of part of the triolein in the membrane occurs more quickly, and, as a consequence, the oscillation period becomes shorter.

The reason why these two types of oscillation appear is still unknown, and shall be the subject of future research.

3.3.2 Temperature dependence of base conductance

As mentioned above, G increased as temperature rose. In order to clarify this tendency quantitatively, the base membrane conductance was investigated over a wide range of temperature. Figure 11 shows an example of the Arrhenius plot for G of an oscillation. The plot fitted well to a straight line in the figure, and for many other oscillations similar results were obtained. The activation energy of G was calculated to be 12.7 kJ mol^{-1} from this Arrhenius plot. The average value of the activation energy for 16 modes of 9 samples investigated was $14.4 \pm 3.3 \text{ kJ mol}^{-1}$. These values are close to the activation energy of the electric conductivity of a 0.5 M KCl aqueous

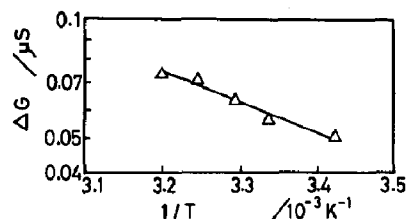


Fig. 12. A typical Arrhenius plot of conductance amplitude for the same sample as that in Fig. 11.

solution, 12.7 kJ mol^{-1} and that of 0.5 M NaCl aqueous salt solution, 13.8 kJ mol^{-1} [14]. From such agreement between the activation energy of G and those of the aqueous solutions separated by the membrane, it can be concluded that the temperature dependence of G is due to the temperature dependence of the mobility of ions in pores of the membrane that do not contribute to the oscillation.

3.3.3 Temperature dependence of conductance amplitude

Conductance amplitude, ΔG , of oscillations also changed with temperature. If the oscillations occurred as a result of opening and closing of a hole through the membrane (channel) as discussed in the previous section, ΔG should correspond to the conductance of the aqueous salt solution that fills the hole, and consequently, temperature dependence of ΔG should resemble that of the aqueous salt solution. Figure 12 shows an Arrhenius plot of ΔG for the same oscillation as that in Fig. 11. The plot fitted well to a straight line. Most of the plots for other oscillations observed also fitted well to an individual straight line for each, except for a few cases where no clear temperature dependence of ΔG was observed. The value calculated for the activation energy from the slope of Fig. 12 is 12.5 kJ mol^{-1} . The average value of the activation energy for 14 modes of 8 samples investigated was $16.4 \pm 3.5 \text{ kJ mol}^{-1}$. Comparing these results with those of base conductance, the average value of the activation energy of ΔG was somewhat larger than that of G , and the variation of values of the activation energy of ΔG was slightly larger than

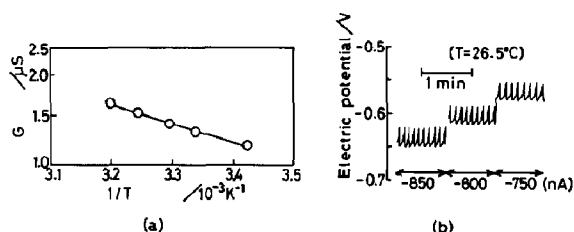


Fig. 11. (a) A typical Arrhenius plot of base conductance. (b) Oscillation curve.

that of G . Nevertheless, values of activation energy of ΔG were also close to those of the bulk aqueous solutions. Such temperature dependence of ΔG is consistent with the explanation given for the oscillations in the previous section. That is to say, the conductance amplitude of an oscillation corresponds to the conductance of the aqueous salt solution that fills the broken part of the membrane, and consequently, the value for the activation energy of the conductance amplitude is close to those of the aqueous salt solutions separated by the membrane. A somewhat larger value of the activation energy may be due to the temperature sensitivity of the area of the broken part. When temperature rises, the area of the broken part may increase because the dynamic viscosity of triolein decreases, and hence, the temperature dependence of ΔG is expected to be slightly larger than that of the salt solution. A change of amplitude of an oscillation itself with time may also have affected the experimental results, because it took at least 1 day to get temperature data.

4. Summary and conclusions

The authors have demonstrated that, by applying an electric current, stable self-sustained oscillations of the electric potential can occur between NaCl and KCl aqueous solutions which are separated by a filter impregnated with triolein. Stable oscillations continued for a long time, and oscillation periods plotted versus base electric potential fitted to a single straight line for each oscillation on a logarithmic scale, even when the membrane conductance gradually decreased while an oscillation continued. It was concluded that the oscillation period was controlled by the base electric potential, not by the electric current applied. The oscillations were explained as rhythmic repetitions of opening and closing of hole(s) in the membrane, due to repetitive breakdown and restoration of some part of the triolein impregnated in the filter. The value of conductance amplitude observed coincided well with this explanation.

When temperature was set higher, base poten-

tial under a certain fixed value of applied current became lower. Stable oscillations were usually obtained in the temperature region between about 0°C and 40°C. Between oscillation period and base potential, two types of relationships were obtained. One was temperature independent, and the other was temperature dependent. In the former case, temperature affected only the base potential. In the latter case, temperature directly affected the oscillations.

Arrhenius plots of base conductance fitted well to a straight line for each of the oscillations observed, and the values of the activation energy calculated from the plots were similar to those of the bulk aqueous salt solutions. Thus it was concluded that the temperature dependence of the base potential is due to the temperature dependence of the mobility of ions in the solutions separated by the membrane. Arrhenius plots of the conductance amplitude of the oscillations were similar to those of the base conductance. This results supports the explanation of the oscillation mechanism mentioned above.

Further studies are now in progress to investigate effects of other factors on the oscillations. What kinds of physical and chemical properties of triolein contribute to stable oscillations should also be investigated. Such investigations will help us to elucidate the oscillation mechanism and to investigate membranes better controllable and more useful.

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