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Nonlinear Behavior and Domain Feature at Oil/Water Interface

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Abstract: Studies on nonlinear behavior at oil/water interface membrane were performed. This system showed rhythmic oscillations and chaos of electrical potential in a given concentration domain. The nonlinear behavior response at the liquid membrane apparently resembled that of biological chemoreceptive membrane. The possibility of developing a new type of chemical sensor with the ability to simulate substance equilibrium in living organisms was suggested in the paper.

Keywords: Nonlinear behavior, oscillations, oil/water interface, domain.

Some of the recent interest in biological pattern formation was sparked by the discovery of spontaneous spatiotemporal pattern formation[1], in a nonliving system, for the Belousov-Zhabotinskii (BZ) reaction. The study of nonliving membrane systems[2-4], such as the artificial systems[5], is much simpler than the biological systems. However the underlying assumption in all of this work is that the nonliving system retains the important features of the biological system.

In this paper, we report a nonlinear phenomenon and their domain feature at an oil/water interface in the presence of phenylalanine (Phe) that is an important constituent of living organisms. The nonlinear behavior response at the liquid membrane apparently resembled that of biological chemoreceptive membrane. The possibility of developing a new type of chemical sensor with the ability to simulate the substance equilibrium in living organisms was suggested according to healthy living organisms, which showed oscillations. From the pattern, a meaningfully studied way is provided to keep substance equilibrium of living organisms.

Experiments were performed in a U-shaped glass tube (15 mm inner diameter), thermostated at 20±1°C. The solutions were added as the following sequence: A solution (6 mL) of Phe in nitrobenzene was placed in the base of the U-shaped glass tube. Then aqueous solutions (5 mL each) were introduced simultaneously into the two arms of the U tube above the organic phase without stirring. The left side of the U-shaped glass tube was an aqueous solution containing cetyltrimethyl ammonium bromide (CTAB) with ethyl alcohol and the right side NaCl aqueous solution. The electrical potential across the liquid membrane was monitored with two platinum Elec-

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Figure 1 Oscillatory curve ofelectrical potential with Phe:1mmol/L, sodium chloride 1mol/L, CTAB 4mmol/L, ethyl alcohol 0.3 mol/L.

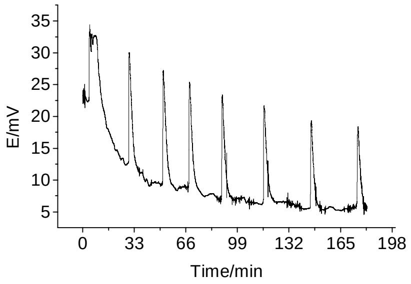
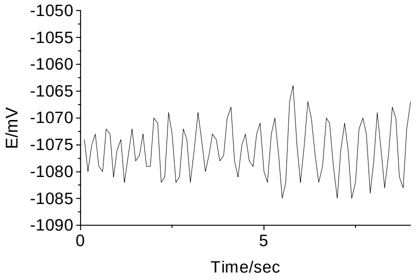


Figure 2 Oscillatory curve of electrical potential with 0.8 mol/L ethyl



Other experimental conditions were the same as for **Figure 1**.

Figure 3 Oscillatory curves of electrical potential with Phe 1 mmol/L, sodium chloride 1 mol/L, ethyl alcohol 0.4 mol/L, CTAB 3

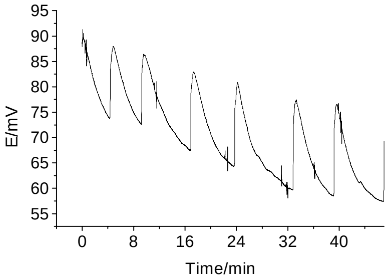
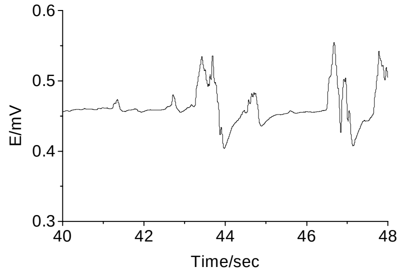


Figure 4 Oscillatory curves of electrical potential with CTAB 5.2 mmol/L.



Other experimental conditions were the same as for **Figure 3**.

Figure 5 The oscillatory domain in the coordinates of the concentration of the CTAB and ethyl alcohol. Rhythmic oscillating domain Ⅰ and chaotic domain Ⅱ.

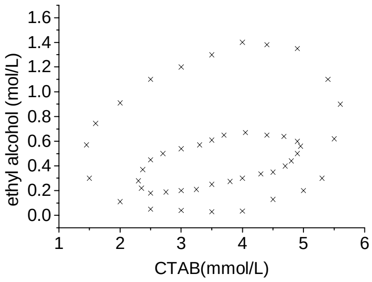
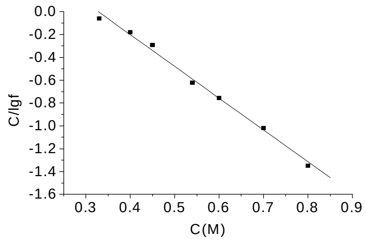


Figure 6 Relationship between C/logf and C.



C is the concentration of an ethyl alcohol, logf is the logarithm of the frequency of oscillation.

trodes situated on either side of the interface. A CHI 832(CHI, USA) was employed to record the changing potential curves.

Throughout the experiments, the concentrations of Phe in nitrobenzene and sodium chloride were fixed at 1 mmol/L and 1 mol/L respectively, for they had not apparent effect on curve potential. However, concentrations of CTAB and ethyl alcohol had remarkable effect on it. Especially, the frequency of oscillations was greatly affected by the concentration of ethyl alcohol. No oscillations were observed in the absence of ethyl alcohol or CTAB.

Figure 1, 2 show the oscillating curves at different concentrations of ethyl alcohol. With the increase of the ethyl alcohol concentration, the frequency of oscillations increased accordingly and rhythmic oscillation became chaotic. But the amplitude of the oscillations was scarcely affected by the concentration of ethyl alcohol.

Figure 3, 4 indicate the oscillating curves at different concentrations of CTAB. When concentrations were decreased, the oscillatory period tended to be shortened and rhythmic oscillations became chaotic. With an increased concentration of CTAB, rhythmic oscillations also became chaotic. Therefore, rhythmic oscillations and chaotic behavior observed had their own concentration domain.

We change the concentrations of the CTAB and ethyl alcohol to detect the oscillatory domain as shown in **Figure 5** (Conditions: 1 mmol/L Phe , 1 mol/L NaCl). The oscillating phenomena were scarcely observed when the concentrations were out of Ⅰ and Ⅱ. The above concentration domains were favorable to a new type of chemical sensor with the ability to simulate the substance equilibrium of living organisms. The simulation may be more useful when the nitrobenzene is replaced by the plant oil and/or surfactant CTAB were replaced by the enzyme.

It is noteworthy that in these experiments, the concentrations of solute were in a state of nonequilibrium. The hydrophobic substances, Phe and CTAB, were dissolved in organic solvent and in water, respectively. The phenomena observed in the two-phase system and in the liquid membrane can be regarded as periodic structure formation at the interface of nonequilibrium state. In the experiment, the frequency of oscillation was sensitive to the ethyl alcohol concentration (C) and the concentration-dependence of the logarithmic response (logf) (**Figure 6**) apparently followed a similar relationship to that of Langumuir's adsorption isotherm:

$$C/(\log f) = 1/\log f_{max} (C + 1/K) \qquad (1)$$

Where C is concentration of ethyl alcohol, $\log f_{max}$ is the maximum response corresponding to the maximum amount of adsorption in langumuir's adsorption isotherm, and K is an apparent equilibrium constant. It was found that the critical concentration to induce the oscillation depended on the hydrophobicity of the ethyl alcohol. The relationship between the critical concentration and the hydrophobicity can be represented by the following equation:

$$\log C_{th} = -a \log P + b \qquad (2)$$

Where C_{th} is the concentration at which the oscillations change sharply, P is the partition coefficient of ethyl alcohol between nitrobenzene and water, a and b are constants.

It is known that ethyl alcohol can decreases the critical micelle concentration (CMC) of amphiphilic molecules[6]. The decrease is due to incorporation of ethyl alcohol into the micelle, which changes the manner of aggregation of the amphilic molecules. The effects of alcohol can induce frequency changes of oscillation.

Acknowledgments

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