Periodic Change Of Viscosity And Density On Oscillating Chemical Reaction

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The Belousov-Zhabotinsky (BZ) reaction is one of the most thoroughly investigated oscillating chemical reactions. The BZ reaction exhibits a wide variety of nonlinear phenomena, e.g., target pattern or spiral pattern in an unstirred shallow solution, and multistability, periodicity, multiperiodicity or deterministic chaos in a stirred solution.

It is obvious that the solution viscosity and density of the BZ reaction plays an important role in the spatial and the temporal patterns. However, the solution viscosity and density of the BZ reaction is not still measured directly. In the previous experiments, the effects of the solution viscosity and density were monitored by the variation of the redox potential and the absorbance. Therefore, it is very important to monitor directly the behavior of the solution viscosity and density on the BZ reaction. In this report, we present the new method to measure the solution viscosity and density of the BZ reaction. In this method, we employ the quartz crystal microbalance (QCM). When the QCM is immersed into a solution, its oscillating frequency shifts depending on the solution viscosity and density, $(\eta_L\rho_L)^{1/2}$(η_L is the absolute viscosity of a solution and ρ_L is the density of a solution) . This characteristic feature enables us to measure the time series of $(\eta_L\rho_L)^{1/2}$of the BZ reaction. In the present experiments, we measure in the stirred system the redox potential and $(\eta_L\rho_L)^{1/2}$ of the BZ reaction at the same time.

We investigated the rhythmic phenomenon in the iron catalyst system and the cerium catalyst system. In the present experiments, the one side of the QCM was sealed with a blank quartz crystal casing (Figure 1b), maintaining it in an air environment, while the other side was contacted with the solution of the BZ reaction. This casing is essential for the frequency stability of the QCM in an ionic solution. The one-face sealed QCM was vertically immersed into the BZ reaction (Figure 1a).

First, we investigated the redox potential and $(\eta_L\rho_L)^{1/2}$ of the iron catalyst system of the BZ reaction. Figure 2, 3 shows the time series of the redox potential (ΔE) and the frequency shift (ΔF, that is, $(\eta_L\rho_L)^{1/2}$), and the ΔE-ΔF phase portrait. It is obvious that ΔF varies in a rhythmic manner, synchronized with the oscillation of ΔE: when the redox potential is high (the ratio of Fe(III) complex to Fe(II) complex is high), $(\eta_L\rho_L)^{1/2}$ is high. Furthermore, it is also found that when ΔE increases drastically, ΔF increases after a delay of about 1 to 3 s. In both ΔE and ΔF, the average period of the oscillation is 35.6 s. The average amplitudes of ΔE and ΔF are 55.5 mV and 221 Hz, respectively. The tendency of the cerium catalyst system was also equal to that of the iron catalyst system.

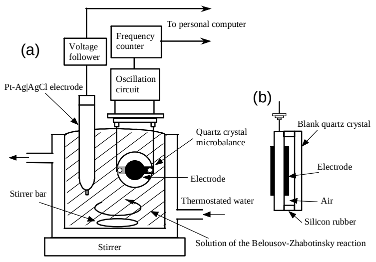

Fig.1. (a) Schematic illustration of the experimental setup. (b) Schematic diagram of the one-face sealed QCM.

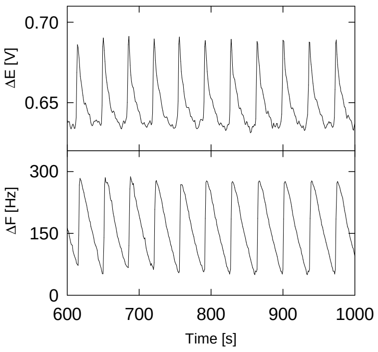

Fig.2. Time series of the iron catalyst system of the BZ reaction..

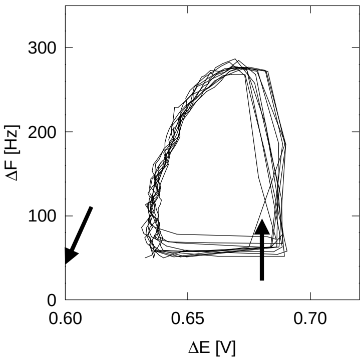

Fig.3. Phase portrait of the iron catalyst system of the BZ reaction.

Reference
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