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T-shaped coincidence detector as a band filter of chemical signal frequency

J. Gorecka[1] and J. Gorecki[2,*]
[1]*Institute of Physics, Polish Academy of Sciences, Al. Lotnikow 36/42, 02-668 Warsaw, Poland*
[2]*Institute of Physical Chemistry, Polish Academy of Sciences, Kasprzaka 44/52, 01-224 Warsaw, Poland*

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The construction of a reactor that works as a bandpass filter for a chemical signal is described and its potential applications are discussed. By a chemical signal we understand a train of concentration pulses propagating in a spatially distributed excitable medium.

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I. INTRODUCTION

Spatially distributed excitable chemical systems can be applied in information processing. Information is related to excitations of such systems, i.e., to peaks of the concentration of a selected reagent that is high in the excited state. According to this assumption, no information is recorded or processed in the stationary state. Information can be loaded by external excitations of the system, it can propagate in the form of concentration pulses and it can be processed in areas of space, where pulses interact. It seems that chemical excitations are responsible for information processing in biological systems, so better understanding of the phenomenon seems to be important [1].

Many recent papers that discuss applications of excitable chemical systems in information processing represent a ''digital'' approach to the problem [2–5]. The binary logic is used so a high concentration of the selected reagent is interpreted as the logical ''true'' state, whereas the state of a low concentration as the logical ''false.'' Thus, a propagating pulse describes a bit of information traveling in space. The information coded in traveling pulses can be processed using properly constructed reactors. Such reactors consist of active areas, where the system is excitable, and passive areas, where some of reactions do not occur, but reagents can diffuse. In experiments, a given pattern of active and passive areas can be created by introducing an immobilized catalyst in a nonhomogeneous way [6–8] or (for a photosensitive reaction) by an inhomogeneous illumination [9]. The chemical information diode, which transmits information in one direction only, was one of the first devices analyzed in numerical simulations and realized in experiments [3,6,7,10]. More complex signal processing reactors, such as logical gates [2,3,11], chemical memory cells [3,4], coincidence detectors [3,5], and switches of the direction of a traveling pulse [5], have also been studied and most of these devices have already been tested experimentally. Using them as building blocks, one can construct reactors that allow for even more complex signal processing, for example, counting the number of pulses arriving at a given point of space [12].

It has been shown recently that a barrier of a passive medium that separates active areas can be used as a transformer of the signal frequency [8,13] because for a properly chosen barrier width, it transmits a given fraction of the arriving pulses. However, calculations performed for ''realistic'' models of excitable chemical medium have shown that the applications of a passive barrier as a filter of chemical signal frequency are quite limited [14].

In this paper, we show how a bandpass filter for a chemical signal can be constructed and tuned to the required frequency range. The distribution of passive and active areas in the filter is generic so it can be applied to any excitable chemical system if the proper widths of passive barriers are selected.

II. THE BANDPASS FILTER FOR A CHEMICAL SIGNAL

The geometry of a bandpass filter is shown in Fig. 1. The black areas correspond to the active regions and the white areas are the passive ones. In the case of the Belousov-Zhabotinsky (BZ) reaction discussed below, the catalyst (ferroin) is immobilized in the active regions, whereas there is no ferroin in the passive ones. The filter is build of two signal channels (B and C) and T-shaped coincidence detector formed by E and F channels. The T-shaped detector [12] does not respond to a single pulse propagating in its signal channel E, but it sends a pulse out when two pulses meet over the detector bar F. The diodes $D1$ and $D2$ block the transmission of signal from E to B and C channels. The difference in lengths of signal channels creates the time shift necessary for signal filtering. In the following, we assume that the C channel is longer than the B channel. The idea of frequency filtering can be explained in the following way.

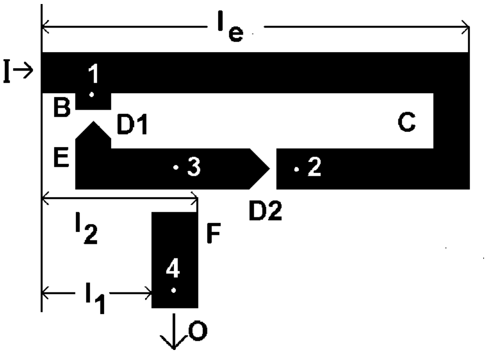

FIG. 1. The geometry of a frequency filter. The black color shows the distribution of the active medium; the passive areas are white.

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[*] Electronic address: gorecki@ichf.edu.pl

Let us consider a periodic train of pulses entering the filter from the input *I*. An arriving pulse splits. One of the created pulses enters the channel *B*, next passes through the diode $D1$ and enters the signal channel, of the T-shaped detector *E*. If it was generated by the first pulse of the train, it dies at the diode $D2$. The second of the separated pulses propagates along the channel *C* and after a time interval, which is determined by the length of this channel, enters *E* through the diode $D2$. If it meets one of the subsequent pulses of the train that propagated through the diode $D1$ above the detector bar *F*, then the detector bar gets excited and the signal in the output channel *O* is generated. If there is no pulse that entered *E* through the diode $D1$ or if the point at which the signals meet is not located just above the detector bar, then there is no output signal. Both types of behavior are presented in Figs. 2(a) and 2(b), where concentration of a selected reagent at a few points on the filter is plotted as a function of time.

Assuming that the pulse velocity is constant, its wave vector in *E* is parallel to the longest site of this channel; and that the same time interval is needed to cross both diodes $D1$ and $D2$, one can show that the detector bar located as shown in Fig. 1 becomes excited if the time interval Δt between consecutive pulses of the train satisfies the relationship

$$\frac{2(l_e-l_2)}{v} \leqslant \Delta t \leqslant \frac{2(l_e-l_1)}{v}, \tag{1}$$

where v is the velocity of a propagating pulse.

The predicted response of chemical frequency filter can be verified by numerical solution of the reaction-diffusion equations. In our calculations we used the Rovinsky-Zhabotinsky (RZ) model of ferroin catalyzed BZ reaction [15,16]. The RZ model uses two variables: *x* and *z*, corresponding to the dimensionless concentrations of the activator $HBrO_2$ and of the oxidized form of catalyst $Fe(phen)_3^{3+}$. In the active regions, the time evolution of the concentrations of *x* and *z* is described by Eqs. (2) and (3):

$$\frac{\partial x}{\partial \tau} = \frac{1}{\epsilon}\left[x(1-x) - \left(2q\alpha\frac{z}{1-z}+\beta\right)\frac{x-\mu}{x+\mu}\right] + \nabla_\rho^2 x, \tag{2}$$

$$\frac{\partial z}{\partial \tau} = x - \alpha\frac{z}{1-z}. \tag{3}$$

In the passive regions, $z=0$ and x evolves according to

$$\frac{\partial x}{\partial \tau} = -\frac{1}{\epsilon}\left[x^2 + \beta\frac{x-\mu}{x+\mu}\right] + \nabla_\rho^2 x. \tag{4}$$

Equations (2)–(4) correspond to a typical experimental situation, in which ferroin is immobilized on a flat membrane, whereas the activator is in the solution and it can diffuse (compare [7,17]). Therefore, free boundary conditions between the active and passive areas are assumed.

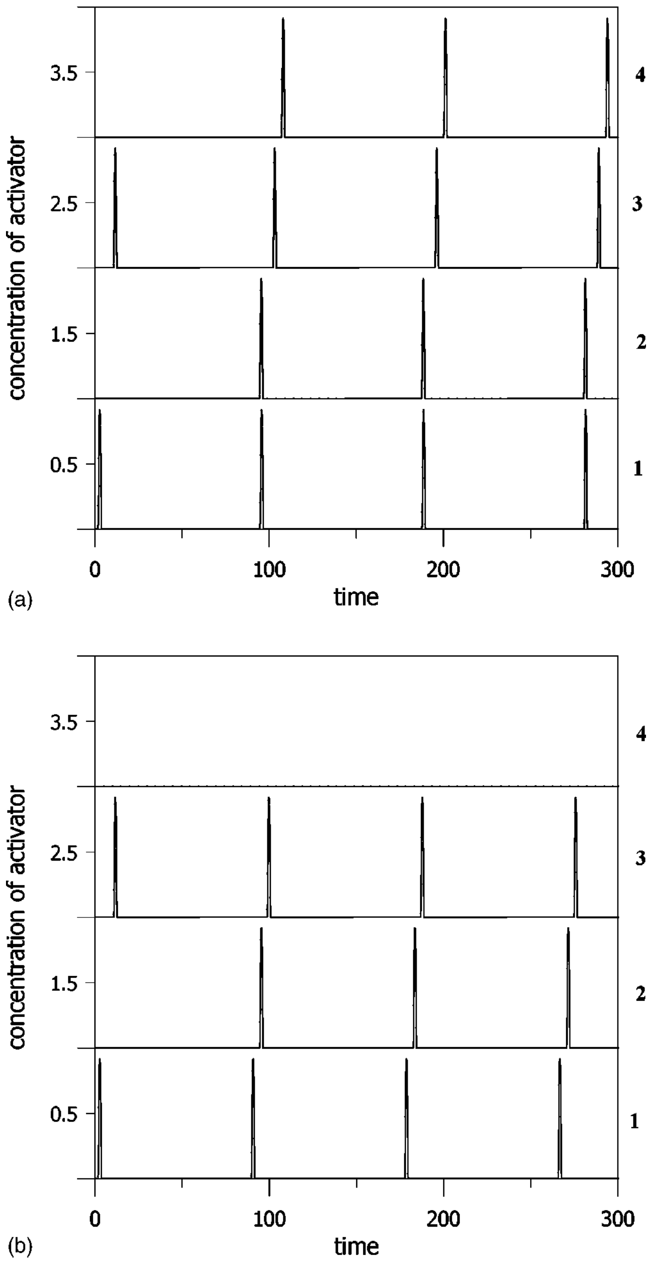

FIG. 2. Curves 1–4 show $x(\tau)$ at points 1–4 marked in Fig. 1. The curves 2, 3, and 4 are shifted up with respect to the scale by 1, 2, and 3 units, respectively. (a) shows the response of the filter to a perturbation with a period $\Delta\tau=93$; the results in (b) are for $\Delta\tau=88$. Both concentration and time are expressed in dimensionless units.

All variables and coefficients in Eqs. (2)–(4) are dimensionless and the real concentrations of $HBrO_2$ and $Fe(phen)_3^{3+}$ (X,Z) are related to (x,z) in the following way:

$$X = \frac{k_1 A}{2k_4}x, \tag{5}$$

$$Z = Cz, \tag{6}$$

where $k_{\pm i}$ denote the rate constants of the corresponding reactions in the Field-Körös-Noyes model [16,15], $A=[HBrO_3]$ and $C=[Fe(phen)_3^{2+}]+[Fe(phen)_3^{3+}]$. The coefficients α, β, μ, and ϵ are defined in Refs. [5,16]. In our numerical calculations, we use the values of α, β, ϵ, μ

equal to 0.017 h_0^{-2}, 0.0017 h_0^{-1}, 0.1176, and 0.00051, respectively [16,15] and $h_0=0.5$ [the system of Eqs. (2) and (3) is excitable for $h_0<0.9899$].

For the values of parameters defined above, the stationary concentrations of x and z in the active area are $x_{sa}=7.283\times10^{-4}$ and $z_{sa}=1.060\times10^{-2}$. The stationary value of x in the passive area is $x_{sp}=5.10\times10^{-4}$.

The relationships between dimensionless time τ and distance ρ used in Eqs. (2)–(4) and the real time t and distance r are the following:

$$t=\frac{k_4C}{k_1^2A^2h_0}\tau, \tag{7}$$

$$r=\sqrt{\frac{k_4C}{h_0}}\frac{1}{k_1A}\sqrt{D_X}\rho, \tag{8}$$

where D_X is the diffusion constant of the activator x. For the parameters chosen,

$$t[\mathrm{sec}]=8.5\tau \tag{9}$$

and for a typical diffusion of reagents in an aqueous solution of $D_X=1\times10^{-5}$ cm^2/sec [7,16,18],

$$r[\mathrm{mm}]=9.218\times10^{-2}\rho. \tag{10}$$

The reaction-diffusion equations for the distribution of active and passive areas shown in Fig. 1 are solved on a grid of 780×160 points. No flow boundary conditions at the grid boundaries are used. The distances between the grid points in both directions are equal to $\Delta\rho=0.3301$ (~0.03 mm). The stripes of the active medium are 30 grid points wide (0.9 mm). The width d_d of passive stripe separating the signal channel of the filter E from the detector channel F is 10 grid points (0.3 mm). The lengths l_e, l_1, and l_2 are equal to 769, 66, and 96 grid points, respectively (23.1, 1.98, and 2.88 mm). The passive area in the middle of the filter is 34 grid points (1.02 mm) wide. The results of Ref. [12] indicate that the distance between the signal and the detector channels in a coincidence detector should be selected with a high precision, so in our calculations the same values of $\Delta\rho$ and d_d as in Ref. [12] have been used.

The response of the filter to input signals generated by increasing the value of x to $x_{ini}=0.1$ at the input of the filter (I) every $\Delta\tau$ time units has been observed. Figures 2(a) and 2(b) show $x(\tau)$ at the points marked as 1, 2, 3, and 4 on Fig. 1 for $\Delta\tau=93$ (~790 sec) and $\Delta\tau=88$ (~748 sec), respectively. In both figures, the bottom line shows concentration at the point 1 located before diode $D1$ (cf. Fig. 1). For both periods of initialization, all pulses enter the filter. The second line from the bottom gives the concentration at the point 2 located at the input of the diode $D2$. For $\Delta\tau=93$, the pulses pass through this diode, enter the signal bar of the detector E, and are observed at the point 3 as the second curve from the top on Fig. 2(a) shows. These pulses face the pulses generated by a subsequent input signal, which entered the signal bar via diode $D1$. The pairs of pulses meet over the detector bar F and excite it. As a result, periodic excitations are observed in the output channel; $x(\tau)$ at the point 4 is represented by the topmost curve in Fig. 2(a). However, if the frequency of the input signal is higher [Fig. 2(b)], then the response of the same filter is different. The peaks observed at the point 3 come from pulses, which entered the signal bar E via the diode $D1$. These meet the pulses that entered E through $D2$ right to the detector bar and no excitation is observed at the point 4 [Fig. 2(a), the topmost curve]. Similarly, there is no excitation of the output channel if the frequency of input pulses is lower, as the pairs of pulses entering E meet on the left to the detector bar (or for yet larger period they propagate independently in the signal bar and die on the diodes). Therefore, the device shown in Fig. 1 works as a band filter for a periodic signal. The mean transmitted frequency f_0 can be calculated as the inverse of the mean time satisfying the relationship (1) and the band width is determined by the length of the detector bar l_2-l_1.

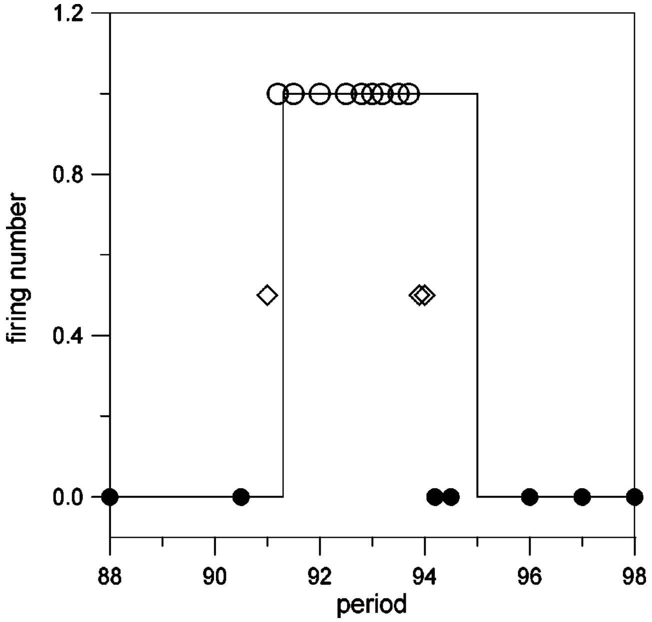

FIG. 3. The firing number as a function of the period of input signal $\Delta\tau$. Symbols represent data obtained form reaction-diffusion equations and the line is an estimation based on relationship (1).

The firing number for a filter can be defined as the ratio of the number of output pulses to the number of input ones. Fig. 3 shows the firing number as a function of $\Delta\tau$ for the considered filter. Open circles correspond to periodic signals for which the filter is fully transparent. The dark dots mark periods of stopped signals. The line presents the firing number estimated from the relationship (1). It is equal to 1 if Eq. (1) is satisfied and it is 0 otherwise. The value of velocity is obtained from a numerical solution of reaction-diffusion equations for a pulse propagating in the active stripe 30 grid points wide. The agreement between the estimation and the exact calculations is quite good, and the differences observed for long periods ($\Delta\tau\sim94$) can be explained by the fact that for the considered geometry a pulse which entered E through $D1$ is not parallel to the channel. It is interesting to note that the firing number can be different from 0 or 1. We observed signals for which every second input pulse is transmitted and the corresponding results are marked with diamonds on Fig. 3. Such a fractional firing number is typical for excitable systems (here *F* channel) perturbed by small periodic excitements and it was discussed in a number of papers (see Refs. [8,13], and references within). The effect, although interesting, is not welcomed in the case of a filter. Fortunately, it can be easily removed if the output signal is filtered again by an identical device.

One can also imagine situations when another type of filter's malfunction occurs. Let us consider a band filter with a short signal bar *E* and a long right channel *C*. Of course, such a device works as a band filter for a low frequency signal and passes signals with frequency around f_0. But if a signal with frequency $2f_0$ arrives, then a pulse propagating through the diode $D2$ may be in coincidence with the next subsequent pulse that entered *E* via $D1$. Thus, the filter can pass the basic frequency as well as a number of harmonic ones. Such a malfunction can be avoided if a long signal bar *E* is used, because in such a case the subsequent pulses annihilate with pulses coming from channel *C* on the right to the detector bar.

III. DISCUSSION

In this paper, we have demonstrated that a simple reactor with geometrical structure shown in Fig. 1 works as a band filter for a periodic chemical signal, because it transmits only those pulses that are separated by certain time intervals. By changing the lengths of signal channels, the position of the detector bar *F*, and its width, the filter can be tuned to the required range of frequencies. But, as a matter of fact, the device is a detector of the time difference between subsequent pulses rather than the filter. It can be used for ‘‘chemical’’ analysis of irregular signals. For example, a number of such devices, tuned to different frequencies, can be applied for direct analysis of a chemical signal. If their outputs are linked to chemical counting devices [12], then the histogram of time intervals between subsequent pulses can be obtained in a purely chemical way.

The presented idea of signal filtering can be generalized, and a chemical reactor that determines whether a selected number of pulses are correlated in time can be easily built. For example, a detector of coincidence between three pulses can be constructed if an additional T-shaped detector is linked with the input channel and with the output of the filter shown on Fig. 1. Now the coincidence between the first two pulses is checked by the first detector and, if such a coincidence occurs, the second detector tests whether these pulses are properly correlated with the third one. If it happens, an output pulse is sent out. Linking more T-shaped detectors together, one can build a device that tests coincidence between any number of pulses.

The signal filtering can be important for systems that code information in frequency of pulses. Such a scenario can be expected in biological systems [1]. The structures discussed above can work as detectors of the fact that the critical excitation has been reached. Of course, the signal filtering can be realized in biological systems in many different ways. For example, the recent paper of Samoilov, Arkin, and Ross [19] demonstrates filtering properties of a network of coupled chemical reactions proceeding in a homogeneous medium.

[1] J.D. Murray, *Mathematical Biology* (Springer, Berlin, 1993).

[2] A. Toth and K. Showalter, J. Chem. Phys. **103**, 2058 (1995).

[3] I.N. Motoike and K. Yoshikawa, Phys. Rev. E **59**, 5354 (1999).

[4] I.N. Motoike, K. Yoshikawa, Y. Iguchi, and S. Nakata, Phys. Rev. E **63**, 036220 (2001).

[5] J. Sielewiesiuk and J. Gorecki, J. Phys. Chem. A **105**, 8189 (2001).

[6] K. Agladze, R.R. Aliev, T. Yamaguchi, and K. Yoshikawa, J. Phys. Chem. **100**, 13895 (1996).

[7] T. Kusumi, T. Yamaguchi, R.R. Aliev, T. Amemiya, T. Ohmori, H. Hashimoto, and K. Yoshikawa, Chem. Phys. Lett. **271**, 355 (1997).

[8] K. Suzuki, T. Yoshinobu, and H. Iwasaki, J. Phys. Chem. A **104**, 6602 (2000).

[9] K. Agladze, A. Toth, T. Ichino, and K. Yoshikawa, J. Phys. Chem. A **104**, 6677 (2000).

[10] A. Toth, D. Horvath, and K. Yoshikawa, Chem. Phys. Lett. **345**, 471 (2001).

[11] O. Steinbock, P. Kettunen, and K. Showalter, J. Phys. Chem. **100**, 18970 (1996).

[12] J. Gorecki, K. Yoshikawa, and Y. Igarashi, J. Phys. Chem. A **107**, 1664 (2003).

[13] J. Sielewiesiuk and J. Gorecki, Phys. Rev. E **66**, 016212 (2002).

[14] J. Sielewiesiuk and J. Gorecki, J. Phys. Chem. A **106**, 4068 (2002).

[15] A.B. Rovinsky and A.M. Zhabotinsky, J. Phys. Chem. **88**, 6081 (1984).

[16] A.B. Rovinsky, J. Phys. Chem. **90**, 217 (1986).

[17] A. Lazar, Z. Noszticzius, H.-D. Försterling, and Z. Nagy-Ungvarai, Physica D **84**, 112 (1995).

[18] R.R. Aliev and A.B. Rovinsky, J. Phys. Chem. **96**, 732 (1992).

[19] M. Samoilov, A. Arkin, and J. Ross, J. Phys. Chem. A **106**, 10205 (2002).