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Belousov-Zhabotinsky "chemical neuron" as a binary and fuzzy logic processor

PIER LUIGI GENTILI[1,2,*], VIKTOR HORVATH[1,3],
VLADIMIR K. VANAG[1,4] AND IRVING R. EPSTEIN[1]

[1]*Department of Chemistry, Brandeis University, Waltham, MA 02454-9110, USA;*
[2]*Department of Chemistry, University of Perugia, 06123 Perugia, Italy;*
[3]*Institute of Chemistry, Department of Analytical Chemistry, L. Eötvös University, P.O. Box 32, H-1518 Budapest 112, Hungary;*
[4]*Department of Biology, M. V. Lomonosov Moscow State University, Moscow 119899, Russia*

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We demonstrate experimentally that the well-known oscillatory Belousov-Zhabotinsky (BZ) reaction can be exploited to process both Boolean and fuzzy logic if the input variables are either the volumes or the phase of addition of pulse-injected solutions of inhibitor (bromide) and activator (silver ion) and the output variable is the oscillation period. Analysis of the relations between the input and the output variables reveals that this oscillating chemical reaction is suitable to process infinite-valued fuzzy logic, and that all fundamental fuzzy logic operators (AND, OR, NOT) can be implemented with it. We discuss the possibility for biological oscillators such as neurons or pacemaker cells to process information using principles of fuzzy logic.

Keywords: Oscillatory Belousov-Zhabotinsky reaction, Boolean logic gates, fuzzy logic systems, pacemaker cells, activator, inhibitor

1 INTRODUCTION

Current digital computers, based upon silicon hardware and programmable software, utilize electrical signals and Boolean logic gates to perform a huge

*Corresponding author: Telephone: +39 075 5855576, FAX: +39 075 5855598; pierluigi.gentili@unipg.it

number of instructions per unit time. A computational speed of eight quadrillion calculations per second (8 petaflops) has been declared to be within reach[1]. Although silicon-based computers are impressively powerful, they still have some technical and computational limitations. Problems involving pattern recognition or decision making in complex situations, for example, are notoriously difficult,[2,3] spurring attempts to devise a new generation of computing machines with novel architectures. The new computing prototypes proposed to date are most often "soups" of chemicals, wherein hardware and software are not completely distinct, because they are embodied, at least partially, in the structures of the molecules that perform the computation[4,5]. The use of "wetware" or "gooware" makes these new computing devices more similar to our brain than to their silicon-based ancestors.

An alternative approach to finding new principles of computation is to abandon Boolean logic and instead apply the fuzzy logic that our brain is thought to utilise[3]. Fuzzy logic is grounded in the theory of fuzzy sets. An item belongs only partially to a fuzzy set, and its degree of membership can assume values between 0 (absence of membership) and 1 (complete membership). An object may belong to more than one set. Based upon fuzzy set theory, a new logic can be processed wherein propositions are not required to be either true or false, but may be true or false to varying degrees. Fuzzy logic aims at modeling the imprecise modes of reasoning that play an essential role in the remarkable human ability to make rational decisions in an environment of uncertainty and imprecision[6]. On the other hand, Boolean logic deals only with certain and objective information. If Boolean logic encodes information in binary digits, fuzzy logic is an infinite-valued logic. Whereas Boolean logic is based upon the law of excluded middle, fuzzy logic breaks that law to some degree[7].

Neurons, the basic elements of our brain, are nonlinear dynamical systems[8]. Therefore, if we want to approach the computational capabilities of the human brain, it is necessary to work with nonlinear dynamical systems, for example, chemical systems that are able to demonstrate oscillatory behaviour. A prototypical example is the Belousov-Zhabotinsky (BZ) reaction,[9,10] the catalytic oxidative bromination of an organic substrate such as malonic acid by bromate in acidic medium[11]. Various metal ions or metallocomplexes, for example cerous ions or ferroin [tris-(1,10-phenantroline)-iron(II)], can serve as the catalyst. In the BZ reaction, the intermediate bromide ion plays a key role[12], since the state of the system depends crucially on its concentration. If its concentration is above a critical value, $[Br^-]_{cr}$, most of the catalyst is in its reduced state (ferroin, for example), while if $[Br^-] < [Br^-]_{cr}$, autocatalytic production of the activator, $HBrO_2$, starts, and the catalyst switches to its oxidized state (e.g., ferriin).

When the BZ reaction is in an oscillatory regime, it can be formally compared to the dynamical behaviour of biological excitable/oscillatory cells (i.e., neurons), which spontaneously depolarize their axon hillock and fire action potentials, often at a regular rate[13]. Although such natural oscillators have their own internal rhythm, external stimuli can alter their timing. In pacemaker cells, for example, information about a stimulus is encoded by changes in the timing of individual action potentials, and it is used to rule proprioception and motor coordination for running, swimming, and flying[14].

The same is true for the BZ reaction. Perturbation of the BZ reaction by different species[15] shifts the phase of oscillations and the time of spike generation, which results from the autocatalysis. The addition of Br^- should lead to a delay in the appearance of a spike, while removal of Br^- should lead to an advance[16]. Perturbations by bromide ions (inhibitor) and/or by silver ions (which quickly remove Br^- from a BZ solution) [16] will play the most important role in the present work.

We show here that the response of the BZ reaction to KBr (inhibitor) and to $AgNO_3$, which we refer to as an "activator" due to its anti-inhibitor action (though the natural, direct activator is $HBrO_2$), applied in different amounts and at different times during the oscillatory cycle, can be exploited to implement certain Boolean logic gates and all fundamental fuzzy logic operators.

To complete this Introduction, we note that BZ reaction waves in spatially extended oscillatory or excitable systems have also been used to build binary logic gates[17-22]. Here, in contrast, we build logical operations from the *spatially uniform* oscillatory BZ reaction.

2 EXPERIMENTAL SECTION

Chemicals. Deionised water and the following analytical grade chemicals are used to prepare the solutions: $NaBrO_3$ (99+%, Acros Chemicals), tris-(1,10-phenantroline)-iron(II) (hereinafter referred to as ferroin) solution (0.025M, Ricca Chemical Company), malonic acid (MA) (99%, Acros Chemicals), H_2SO_4 (10N, Fisher), KBr (99+%, Janssen Chimica), $AgNO_3$ (100%, Fisher), $HClO_4$ (70%, Fisher), K_2SO_4 (99+%, Acros Chemicals).

Experimental setup. The BZ reaction is carried out in a continuously fed stirred tank reactor (CSTR) consisting of a beaker sealed by a homemade Teflon stopper, and having a volume of 30 mL. Two stock solutions of BZ reactants (sodium bromate, malonic acid, sulphuric acid, and ferroin) feed the CSTR *via* a peristaltic pump (Gilson Minipuls 3). The flow rates of the two channels are equal, with a deviation from their average of less than 1%. The residence time of the CSTR is 13 min. The volume of the solution, which is stirred at 500 rpm with a magnetic stirrer (Scinics co., Multi-Stirrer MC 301), is kept constant by continuous removal of the excess reaction mixture through a hole in the Teflon stopper using an aspirator pump (Oakton WP-15). All experiments are performed at room temperature (293 ± 1K). The reaction in the CSTR is monitored with a Pt electrode (Pt, Radiometer M241Pt). As a reference electrode, we employ a silver/silver chloride/potassium chloride electrode (REF, Radiometer REF321) connected to the reaction mixture through a salt (saturated K_2SO_4) bridge (Radiometer AL100). The electrodes are connected to a potentiometer (Oakton pH 510). The analog output of the potentiometer is connected to the analog input of a data acquisition board (National Instruments, NI-6531 USB). Data are collected at a constant rate of 2 samples per second.

Perturbations of the BZ limit cycle are performed by introducing solutions of activator or inhibitor into the CSTR in amounts controlled by two normally closed electronic solenoid valves (Takasago Electric, Japan STV-2-1/4UKG). To open the valves, we use the digital 5V TTL output of the data acquisition board. The valves require a 10 V potential and high current to be opened, while the TTL output signals of our data acquisition board give lower current and smaller voltage. Therefore, we employ reed relays (COTO-9007-05-0000-1032) grouped on a relay board with one reed relay for each valve, operated independently, to switch on and off the constant output of the power supply unit (Extech 382260). The reed relays have a very short switching time, 0.5 ms, and are operated under low current (supplied by the TTL output signals from the data acquisition board). We find a linear response between the amount of solution discharged and the duration of the open state of the solenoid valve, with a lower limit of 25 ms. The opening times of the valves are in the range 100-900 ms, corresponding to 10 μL-90 μL of inhibitor or activator solution. The inhibitor released into the CSTR is KBr (0.0155 M) in H_2SO_4, while the activator is $AgNO_3$ (0.005 M) in $HClO_4$. The concentration of H_2SO_4 in the inhibitor solution, 0.3 M, is set equal to the concentration of H_2SO_4 in the CSTR in order to avoid dilution of H^+, since the period of oscillation is sensitive to even small changes in $[H^+]$. For the same reason, to the solution of activator, we add perchloric acid (0.55 M), after verifying it does not affect the oscillating BZ reaction. We choose $HClO_4$ instead of sulphuric acid to avoid possible precipitation of silver sulphate. The concentration of H^+ in the activator solution is roughly the same as that in the CSTR.

Determinations of the period variation, $\Delta T = T_{pert} - T_0$ (where T_{pert} and T_0 are the periods of the perturbed and natural oscillations, respectively) are repeated at least twice. The uncertainty in the ΔT values is ± 0.2 s.

Computational methods. To build fuzzy logic systems (FLSs) using the methods[23-25] of Mamdani and Sugeno, we employ the Fuzzy Logic Toolbox for Use with MatLab (*MathWorks*).

3 RESULTS

We investigate perturbations of the BZ reaction by Br^- and Ag^+ ions with the following initial BZ concentrations in a CSTR: $[NaBrO_3]$ = 0.25 M, [MA] = 0.3 M, [ferroin] = 1 mM, and $[H_2SO_4]$ = 0.3 M, and residence time k_0^{-1} = 13 min. Under these conditions, the BZ reaction exhibits oscillations

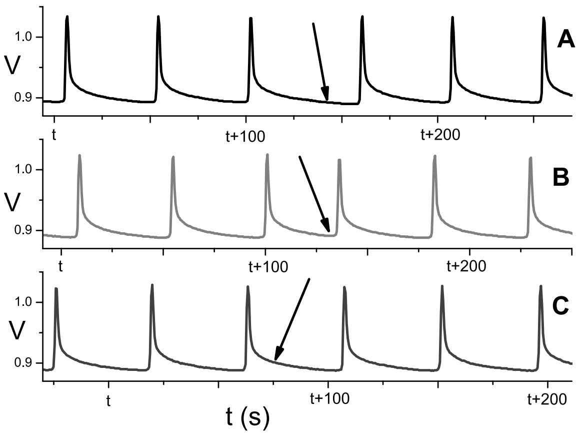

FIGURE 1
Effect of addition of KBr and $AgNO_3$ solutions on the period of oscillations measured by the potential (V) of a Pt electrode. Addition of (A) 60 μL KBr solution at phase $\phi = t/T_0 = 0.6$; (B) 40 μL of $AgNO_3$ solution at phase $\phi = 0.6$; (C) 10 μL of $AgNO_3$ solution at phase 0.2. The instants of addition are indicated by arrows.

with a period of about 45 s (see Figure 1). In Figure 1A, we show that addition of inhibitor increases the period of oscillation. The moment when the injection of either inhibitor or activator occurs will be characterized by the phase $\phi = \tau/T_0$, where τ is the "time delay", i.e., the time since the most recent spike recorded by the Pt electrode. In Figure 1A, for example, the period increases from 48 s to 58 s.

In Figure 1B, we demonstrate that addition of Ag^+ at $\phi = 0.6$ decreases the period, from 46 s to 35 s in the present case, because the silver ion irreversibly removes bromide, forming nano/micro-crystals of AgBr. However, the role played by $AgNO_3$ is not always what one would anticipate for an "activator". When Ag^+ is injected in small quantities and at low phase, it generates a slight *lengthening* rather a shortening of the period. An example of such behaviour is shown in Figure 1C, where the period increases from 43.2 s to 44.1 s upon addition of a small quantity of $AgNO_3$ at $\phi = 0.2$.

This "dual" effect of $AgNO_3$ has also been observed by others[16] in the cerium-catalysed BZ reaction in a closed system. Two explanatory hypotheses have been proposed. The first is based on the buffering effect of bromo-complexes of Ag^+,[26] whereas the other invokes the formation of cerium-bromide complexes. Since our experiments employ ferroin rather than cerium as the catalyst, we adopt the first hypothesis as the more likely explanation of the observed behaviour.

A plot of the dependence of the period variation on the amount of KBr or $AgNO_3$ added at different phases is shown in Figure 2. We express these

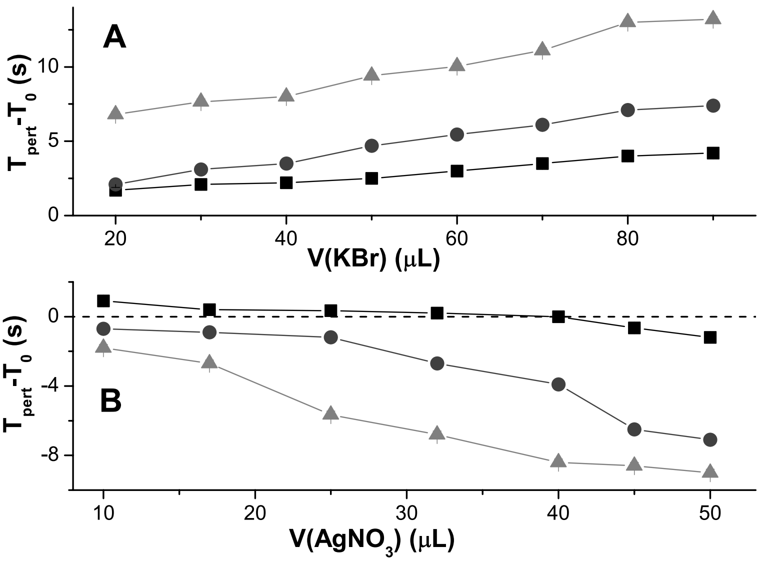

FIGURE 2
Dependence of $\Delta T = T_{pert} - T_0$ on (A) V(KBr), the volume KBr solution, and (B) $V(AgNO_3)$, the volume of $AgNO_3$ solution injected at different phases $\phi = 0.2$ (black squares), 0.4 (gray circles), and 0.6 (light gray triangles).

amounts in terms of the volume of injected solution, V(KBr) or $V(AgNO_3)$, though these units are easily converted to concentrations. On examining plot (A) of Figure 2, it is clear that ΔT always assumes positive values for KBr perturbation and increases with the quantity injected. The relations determined at several phases of addition are smooth, almost linear, and their slopes increase with the phase of addition. Plot (B) of Figure 2 demonstrates the “dual” effect exerted by $AgNO_3$ on ΔT. When Ag^+ is injected at low phase ($\phi = 0.2$, black squares), ΔT assumes positive and negative values for small and large values of $V(AgNO_3)$, respectively. On the other hand, when $AgNO_3$ is injected at higher phases, it always reduces the period, behaving as a pure activator. At all phases, the slopes of the ΔT *vs.* $V(AgNO_3)$ curves are negative and become steeper as ϕ increases. As $V(AgNO_3)$ tends to zero, ΔT must also approach zero. Therefore, the black curve in panel B ($\phi = 0.2$) must have a maximum, which we did not attempt to locate, at small $V(AgNO_3)$.

These nonlinear dependences of ΔT on $V(AgNO_3)$, V(KBr) and the phase of the injection of either inhibitor, ϕ(KBr), or activator, $\phi(AgNO_3)$, allow us to build different logic gates.

Binary logic. In Table 1, we show how to construct the logic function OR. We select $\phi(\text{KBr}) = \phi(AgNO_3) = 0.2$, assign V(KBr) and $V(AgNO_3)$ as the inputs and ΔT as the logical output, under the convention that the output is 0

INPUTS		OUTPUT
***V*(KBr)**	$V(AgNO_3)$	**Δ*T***
0 *(0 μL)*	**0** *(0 μL)*	**0** *(0 s)*
1 *(30 μL)*	**0** *(0 μL)*	**1** *(2.1 s)*
0 *(0 μL)*	**1** *(10 μL)*	**1** *(0.9 s)*
1 *(30 μL)*	**1** *(10 μL)*	**1** *(1.8 s)*

TABLE 1
Truth table for the binary OR logic function. The volumes of the KBr and $AgNO_3$ solutions are the inputs, and the period variation (ΔT) is the output. The chemicals are injected at phase 0.2. Positive logic conventions are used for all variables: the inputs are 0 when the volumes injected are zero, and 1 when they are > 0. The output is 0 when $\Delta T \leq 0$ s, and 1 when $\Delta T > 0$ s. The margins of validity of these conventions are fixed by the accuracy of determination of variables involved.

when $\Delta T \leq 0$, and 1 when $\Delta T > 0$. If $V(AgNO_3)$ is chosen small enough (10 μL) to give a positive ΔT, the function OR is obtained.

On the other hand, if we change $\phi(KBr) = \phi(AgNO_3)$ from 0.2 to 0.6, without altering our logic conventions for the variables, the logic function [TRUE whenever *V*(KBr) is TRUE] can be implemented (see Table 2), because $AgNO_3$ always behaves as an activator when added at $\varphi = 0.6$ (specifically, it decreases ΔT).

If $\phi(KBr)$ and $\phi(AgNO_3)$ are chosen as inputs and ΔT as output, it is also possible to implement the converse non-implication [TRUE if not $\phi(KBr)$ but $\phi(AgNO_3)$]. This operator requires positive logic conventions for the inputs [INPUT = 0 if $\phi = 0.2$ and INPUT = 1 if $\phi = 0.6$] and a negative logic convention for the output (see Table 3). Obviously, if we adopt a positive logic convention for the output (last column in Table 3), we obtain the reverse logic function, which is called "then/if" or "converse implication".

INPUTS		OUTPUT
***V*(KBr)**	$V(AgNO_3)$	**Δ*T***
0 *(0 μL)*	**0** *(0 μL)*	**0** *(0 s)*
1 *(30 μL)*	**0** *(0 μL)*	**1** *(7.7 s)*
0 *(0 μL)*	**1** *(10 μL)*	**0** *(-1.8 s)*
1 *(30 μL)*	**1** *(10 μL)*	**1** *(7.7 s)*

TABLE 2
Truth table for the binary logic function [TRUE whenever *V*(KBr) is TRUE]. The volumes of the KBr and $AgNO_3$ solutions are the inputs, and the period variation (ΔT) is the output. The chemicals are injected at phase 0.6. Positive logic conventions are fixed for all the variables as in Table 1.

INPUTS		OUTPUT_1	OUTPUT_2
ϕ(KBr)	ϕ($AgNO_3$)	**Δ*T***	**Δ*T***
0 *(0.2)*	**0** *(0.2)*	**0** *(3.6 s)*	**1** *(3.6 s)*
1 *(0.6)*	**0** *(0.2)*	**0** *(14.3 s)*	**1** *(14.3 s)*
0 *(0.2)*	**1** *(0.6)*	**1** *(-1.5 s)*	**0** *(-1.5 s)*
1 *(0.6)*	**1** *(0.6)*	**0** *(13.1 s)*	**1** *(13.1 s)*

TABLE 3
Truth table for the [True if not ϕ(KBr) but ϕ($AgNO_3$)] binary logic function. The phases of injection of the KBr and $AgNO_3$ solutions are the inputs, and the period variation (ΔT) is the output. Positive logic conventions are used for the inputs (0 for $\phi = 0.2$, 1 for $\phi = 0.6$) and output_2, whereas a negative logic convention is used for output_1: the output is 0 when $\Delta T \geq 0$, and 1 when $\Delta T < 0$.

It is clear that the oscillating BZ reaction can be used as a configurable Boolean logic element, if we select different combinations of *V*(KBr), *V*($AgNO_3$), ϕ(KBr), and ϕ($AgNO_3$) as the two inputs and ΔT as the output.

It is worth noting that all binary logic gates presented here encode information in the time domain. Note also that the logic gates presented have relatively short reset times, because under our experimental conditions the BZ reaction recovers its initial period in one full period (i.e. a few tens of seconds) after a perturbed oscillation.

Fuzzy logic. To implement fuzzy logic, it is desirable to have smooth dependences of the output, e.g., ΔT, on the inputs, in our case *V*(KBr) and *V*($AgNO_3$) or ϕ(KBr) and ϕ($AgNO_3$)[27,28]. Figure 2 and the ΔT values as a function of several combinations of *V*(KBr), *V*($AgNO_3$), ϕ(KBr), and ϕ($AgNO_3$) reported in Table 4 demonstrate that these dependences are indeed smooth.

With Mamdani's method,[24,29] implementing fuzzy logic operators requires us to build fuzzy logic systems by following a three-step procedure (more mathematical details can be found elsewhere[30]). First, all variables (inputs and output) must be partitioned into fuzzy sets, and a label must be assigned to each set[31]. In Figure 3, we show how inputs *V*(KBr), *V*($AgNO_3$), and φ and output ΔT can be partitioned. Three fuzzy sets, with Low (L), Medium (M), and High (H) as labels, are fixed for the phase ϕ, whereas four fuzzy sets are designated for *V*(KBr), *V*($AgNO_3$): the fuzzy singleton Null (N) and three other sets having Low (L), Medium (M), and High (H) as labels. Note that, because the accuracy of our volume measurements is 1-2 μL, the N fuzzy set actually has finite measure, since very small injection volumes have nonzero degree of membership in N. The output variable is partitioned into five fuzzy sets, with Very Low (VL), Low (L), Medium (M), High (H), and Very High (VH) as labels.

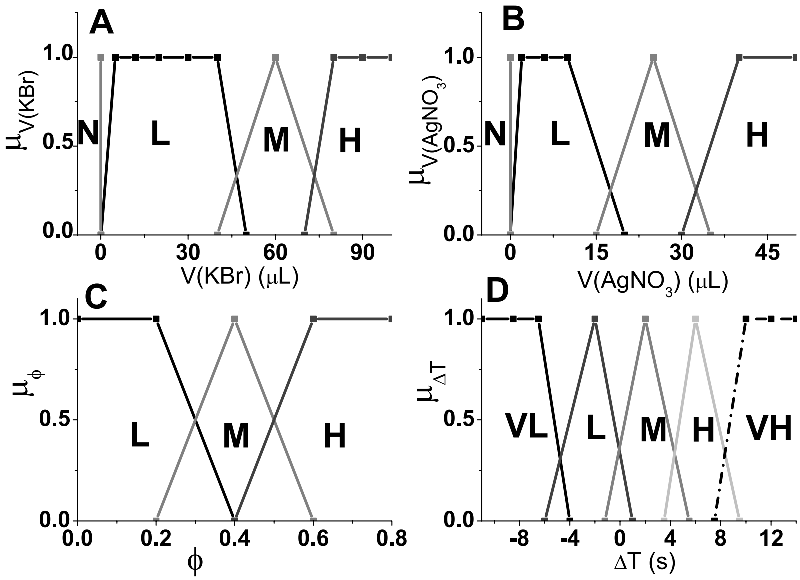

FIGURE 3
Partitions in fuzzy sets of (A) the volume of injected KBr solution [*V*(KBr)]; (B) the volume of injected $AgNO_3$ solution [*V*($AgNO_3$)]; (C) the phase (φ) of addition of either KBr or $AgNO_3$ solution; (D) the period variation ΔT. Ordinates represent the membership function (μ) values for the fuzzy sets. A label is assigned to each fuzzy set: N stands for Null, VL for Very Low, L for Low, M for Medium, H for High, and VH for Very High.

The second step in Mamdani's method entails assigning every numerical value of a variable to a fuzzy set of that variable. An element belongs to all the fuzzy sets, but with different degrees of membership, μ, that range between 0 and 1. An element is assigned to the fuzzy set in which it has the largest degree of membership.

As in the case of the binary logic gates, we can exploit either pair, *V*(KBr) and *V*($AgNO_3$), or φ(KBr) and φ($AgNO_3$), as inputs and ΔT as output. For instance, by using *V*(KBr) and *V*($AgNO_3$) as inputs and ΔT as output, three fuzzy logic systems can be built, based upon the data presented in Table 4 and Fig. 2. They are shown in Fig. 4. The surfaces in Fig. 4 have shaded patches representing the output's fuzzy sets. All three surfaces in Fig. 4 are smooth and hence are ideal for implementation of fuzzy logic operators, the final step of Mamdani's method. This step consists of formulating rules in the form of "IF…, THEN…" statements correlating the input's fuzzy sets to the output's fuzzy sets. If there are two (as in our case) or more inputs, their fuzzy sets will be connected through the AND, OR, and NOT operators.

The plot in Figure 4a is symmetric, because both bromide anion and silver cation play the role of "inhibitor" if injected at low φ (see Figure 2). The rules

ϕ(KBr)	V(KBr) μL	ϕ($AgNO_3$)	*V*($AgNO_3$) μL	Δ*T* (s)
0.2	30	0.2	10	1.75
0.2	30	0.2	25	1.9
0.2	30	0.2	40	1.05
0.2	60	0.2	10	2.6
0.2	90	0.2	10	3.25
0.2	60	0.2	25	1.85
0.2	60	0.2	40	2.8
0.2	90	0.2	25	3.0
0.2	90	0.2	40	3.6
0.4	30	0.4	10	3.15
0.4	30	0.4	25	3.2
0.4	30	0.4	40	2.8
0.4	60	0.4	10	5.8
0.4	90	0.4	10	7.35
0.4	60	0.4	25	5.85
0.4	60	0.4	40	5.35
0.4	90	0.4	25	7.85
0.4	90	0.4	40	7.95
0.6	30	0.6	10	7.7
0.6	30	0.6	25	7.05
0.6	30	0.6	40	6.4
0.6	60	0.6	10	11.15
0.6	90	0.6	10	13.55
0.6	60	0.6	25	10.7
0.6	60	0.6	40	10.6
0.6	90	0.6	25	13.25
0.6	90	0.6	40	13.1
0.2	90	0.2	40	3.6
0.2	90	0.4	40	3.45
0.2	90	0.6	40	-1.5
0.4	90	0.2	40	7.9
0.6	90	0.2	40	14.3
0.4	90	0.6	40	6.9
0.6	90	0.4	40	14.4
0.2	60	0.4	25	1.9
0.2	60	0.6	25	0.05
0.4	60	0.2	25	7.0

0.6	60	0.2	25	11.65
0.4	60	0.6	25	5.2
0.6	60	0.4	25	11.8
0.2	30	0.4	10	1.65
0.2	30	0.6	10	0.35
0.4	30	0.2	10	4.5
0.6	30	0.2	10	8.0
0.4	30	0.6	10	3.5
0.6	30	0.4	10	7.9

TABLE 4
Period variation, ΔT, as a function of the volumes and phases of KBr and $AgNO_3$ solutions injected into the oscillating BZ reaction in a CSTR.

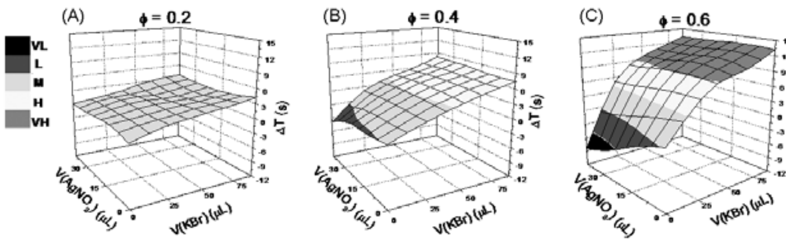

FIGURE 4
Three-dimensional representations of three fuzzy logic systems based on V(KBr) and $V(AgNO_3)$ as inputs and ΔT as output at three different phases of chemical injection: $\phi = 0.2$ (A), 0.4 (B), 0.6 (C). The output fuzzy sets are represented by shadings as indicated in the sidebar at the left.

that can be formulated for this system involve the AND and OR operators. They are of the type:

$$\text{IF } V(\text{KBr}) \text{ is L OR } V(\text{AgNO}_3) \text{ is L, THEN } \Delta T \text{ is M} \quad (1)$$

$$\text{IF } V(\text{KBr}) \text{ is L AND } V(\text{AgNO}_3) \text{ is L, THEN } \Delta T \text{ is M} \quad (2)$$

Note that in eq. (1), while some L values of the inputs can generate ΔT values that have nonzero membership in the L fuzzy set of ΔT, the output is nevertheless M, because these ΔT values have a higher degree of membership in the M set. The complete collection of rules (rules matrix) for the case of ϕ = 0.2 is reported in the left third of Table 5.

When $\phi = 0.4$ (Fig. 4b) or 0.6 (Fig. 4c), the surfaces are asymmetric, and the rules that can be formulated involve the AND and NOT operators (see Table 5 for

4a					4b					4c				
V(KBr)	$V(AgNO_3)$				V(KBr)	$V(AgNO_3)$				V(KBr)	$V(AgNO_3)$			
	L	M	H	N		L	M	H	N		L	M	H	N
L	*M*	*M*	*M*	*M*	L	*M*	*M*	*M*	*M*	L	*H*	*H*	*H*	*H*
M	*M*	*M*	*M*	*M*	M	*H*	*H*	*H*	*H*	M	*VH*	*VH*	*VH*	*VH*
H	*M*	*M*	*M*	*M*	H	*H*	*H*	*H*	*H*	H	*VH*	*VH*	*VH*	*VH*
N	*M*	*M*	*M*	*M*	N	*L*	*L*	*L*	*M*	N	*L*	*VL*	*VL*	*M*

TABLE 5
Rules matrices for the three fuzzy logic systems based on V(KBr) and $V(AgNO_3)$ as inputs and ΔT as output and determined at three different phases of chemical injection: 4a is at $\phi = 0.2$, 4b at $\phi = 0.4$, 4c is at $\phi = 0.6$. The fuzzy sets of ΔT are indicated in italics.

the complete collection of rules for the two FLSs). An example of a rule using the NOT and AND operators, valid for the FLS represented in Figure 4b, is:

$$\text{IF V(AgNO}_3\text{) is NOT N AND } V\text{(KBr) is N, THEN } \Delta T \text{ is L} \qquad (3)$$

Other FLSs can be built by using ϕ(KBr) and $\phi(AgNO_3)$ as inputs and ΔT as output. Three of these are depicted in Figure 5 and are relative to different combinations of the volumes of the KBr and $AgNO_3$ solutions (see data in Table 4). These surfaces are not symmetric. Therefore the respective FLSs will have rules involving the AND and NOT operators, but not the OR operator. The complete collection of rules is given in Table 6. For instance, the FLS of Figure 5a involves a rule such as:

$$\text{IF } \phi\text{(AgNO}_3\text{) is NOT H AND } \phi\text{(KBr) is L, THEN } \Delta T \text{ is M} \qquad (4)$$

whereas the FLS of Figure 5c involves a rule such as:

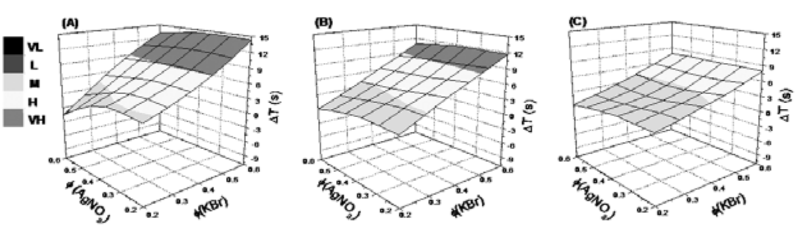

FIGURE 5
Three-dimensional representations of three fuzzy logic systems having the phases of injection of KBr and $AgNO_3$ solutions [ϕ(KBr) and $\phi(AgNO_3)$, respectively] as inputs and ΔT as output. The volumes of the KBr and $AgNO_3$ solutions are 90 μL and 40 μL in (A), 60 μL and 25 μL in (B), 30 μL and 10 μL in (C), respectively. The output fuzzy sets are represented by shadings as indicated in the sidebar at the left.

	5a				5b				5c		
ϕ(KBr)	$\phi(AgNO_3)$			ϕ(KBr)	$\phi(AgNO_3)$			ϕ(KBr)	$\phi(AgNO_3)$		
	L	M	H		L	M	H		L	M	H
L	*M*	*M*	*L*	L	*M*	*M*	*M*	L	*M*	*M*	*M*
M	*H*	*H*	*H*	M	*H*	*H*	*H*	M	*H*	*M*	*M*
H	*VH*	*VH*	*VH*	H	*VH*	*VH*	*VH*	H	*H*	*H*	*H*

TABLE 6
Rules matrices for the fuzzy logic systems having phases of injection of KBr and $AgNO_3$ solutions [ϕ(KBr) and $\phi(AgNO_3)$, respectively] as inputs and ΔT as output. The volumes of the KBr and $AgNO_3$ solutions are 90 µL and 40 µL in (5a), 60 µL and 25 µL in (5b), 30 µL and 10 µL in (5c).

$$\text{IF } \phi(AgNO_3) \text{ is H AND } \phi(\text{KBr}) \text{ is NOT H, THEN } \Delta T \text{ is M} \quad (5)$$

4 DISCUSSION AND CONCLUSIONS

In this work we have demonstrated that smooth variations of input variables such as the volume and the instant of injection of activator and inhibitor solutions determine smooth modifications of the output variable, $\Delta T = (T_{pert} - T_0)$. Therefore, the relations between either V(KBr) and $V(AgNO_3)$ or ϕ(KBr) and $\phi(AgNO_3)$ as inputs and ΔT as output are suitable for implementing the fundamental fuzzy logic operators. These dependences of ΔT on injected inhibitor and activator make the oscillatory BZ reaction the second example of a chemical system in which all the fundamental fuzzy logic operators, AND, OR, NOT, can be implemented, allowing any other complex logic function to be processed. The first such system was based on the chromogenic properties of a spirooxazine[32]. There are previous examples of chemical implementation of fuzzy logic, but these allow implementation only of the AND logic function[33-35]. If we compare the properties of the chromogenic spirooxazine in a solution in a cuvette with those of the oscillating BZ reaction in a CSTR, we conclude that the latter has some advantages over the former: (*i*) it has a faster reset time, requiring a few tens of seconds or less rather than hundreds of seconds; (*ii*) it operates in an open system wherein the content of the solution is constantly renewed and the products of the computation do not accumulate, which allows the use of different oscillatory cycles for different logical operations.

Are nonlinear oscillatory (limit cycle) dynamics in general, and oscillatory living cells in particular, associated with fuzzy logic? Although the detailed workings of pacemakers or other oscillatory cells are quite different from those of an oscillating BZ reaction, the similarities in their dynamical behaviour suggest that a pacemaker cell could process fuzzy logic in much the same fashion as the BZ reaction does. If so, it may well be that our nervous system utilises the principles of fuzzy logic[27] in its functioning.

Further progress in this research will require studying the behaviour of *networks* of such "chemical neurons", e.g., two or more BZ reactions carried out in separate CSTRs and coupled by pulses of activator and inhibitor species[36]. The purpose of these studies will be to assess what kind of logic can be processed and what are the computational capabilities of an ensemble of coupled nonlinear chemical oscillators.

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NOMENCLATURE SECTION

BZ = Belousov-Zhabotinsky
$[Br^-]$ = concentration of bromide ion
CSTR = continuously fed stirred tank reactor
ΔT= period variation
T_{pert} = period of perturbed oscillation
T_0 = period of natural oscillation
FLS = Fuzzy Logic System
ϕ = phase of injection
τ = time delay
V = volume injected.
μ = degree of membership function
N, VL, L, M, H, VH = linguistic labels for Null, Very Low, Low, Medium, High, Very High fuzzy sets, respectively.

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