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Experimental logical gates in a reaction-diffusion medium: The XOR gate and beyond

Andrew Adamatzky[1] and Benjamin De Lacy Costello[2]
[1]*Faculty of Computing, Engineering and Mathematical Sciences, University of the West of England, Bristol BS16 1QY, United Kingdom*
[2]*Faculty of Applied Sciences, University of the West of England, Bristol BS16 1QY, United Kingdom*

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We exploit the particulars of diffusive wave front interactions in certain types of two-reactant reaction-diffusion medium to construct a laboratory prototype of an XOR gate. In the design, the values of the logic variables are represented by the presence or absence of a precipitate, ''wires'' are constructed of a substrate loaded gel, and the computation is based on diffusive wave dynamics. We also discuss implementation of AND gate and study a three-valued composition, derived from the gate dynamic, and discuss possible logics that could be derived from this composition.

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I. INTRODUCTION

Reaction-diffusion processes in chemical media became a hot subject of investigation in the context of unconventional computing devices after the publication of the seminal paper by Kuhnert, Agladze, and Krinsky [1], who demonstrated that it was possible to implement some basic operations of image processing using the light-sensitive Belousov-Zhabotinsky reaction. During the last decade experimental prototypes of reaction-diffusion computing devices were fabricated and applied to solve various problems of computer science, including image processing [1–4], pattern recognition [2,5–9], path planning [10–13] and many more (see overview in Ref. [14]).

Are similar reaction and diffusion based systems constructed in chemical media capable of universal computation? Yes, and this is demonstrated via the implementation of logical gates [5,7,13,15,16] and the simulation of finite automata networks [17–20] in chemical and biochemical systems. There are two ways to implement a logical operation in chemical systems: integral dynamic based and geometry, or wave interaction, based approaches.

The first approach is to exploit chemical kinetics in mass-coupled networks of chemical reactors [5,7,17,20]. Thus, e.g., Blittersdorf *et al.* [15] built Boolean logical gates from a network of three chemical reactors, where two of the reactors represent input variables and the third reactor is an output. Binary states of the reactors equate to the level of acidity in the reagent solutions. The computation is performed by setting up pH levels in the input reactor, adjusting the flow rate between the reactors, and measuring pH in the output reactor. In a similar way an exclusive OR gate was realized in a controlled enzymatic dynamic by Zauner and Conrad [17,20]; this can be seen as a combination of mass-coupled computation and macro-molecular computing, as, e.g., realization of XOR operation in self-assembling of DNA molecules [21].

The second approach, which is adopted in this paper, is based on wave front dynamics in a geometrically constrained media. A logical value is represented by the presence or absence of a wave front at a specified location, a computation is performed when two or more wave fronts interact with other. For example a threshold of state switching, is employed in the design of chemical wave logical gates in Refs. [13,16]; there the computation is based on the fact that an excitable wave in the Belousov-Zhabotinsky reaction is generated only when a critical nucleation size is exceeded. Thus, two excitation waves (input variables) facilitate formation of the third wave (output variable). Clearly, one can also design an XOR gate in some excitable media, where colliding waves annihilate one another, e.g. in a neural dendritic tree [22].

The paper is structured as follows. In Sec. II we describe an experimental setup of the reaction-diffusion chemical processor. An implementation of exclusive OR logical gate is discussed in Sec. III. Some ideas on how to implement AND gate using instabilities in reaction-diffusion system are discussed in Sec. IV. A three-valued binary composition, derived from extended interpretation of spatial states of the chemical processor, is introduced in Sec. V. The paper concludes by Sec. VI, where results on logical computation are considered in a wider context of general purpose computing.

II. PALLADIUM PROCESSOR

Our approach exploits the fact that in a reaction-diffusion processor two diffusive waves of the same reactant but initiated in separate locations form a distinctive pattern when they interact (collide) with each other. The methodology for the production of such processors is originated in Refs. [23,24] and it gets a sophisticated development in recent papers [2,10,25].

A reaction-diffusion logical gate based on palladium chloride is designed as follows. A gel (2% agar or agarose by weight) containing palladium chloride (0.2% by weight) is prepared and allowed to set in Petri dishes or on acetate sheets to a thickness of 1 mm. Geometric structures, including T-shaped chambers and gate couples are cut from the gel film. The gel film forms an active substrate. The values of logical variables are represented by planar substrates soaked in a reactant solution of potassium iodide (saturated at 20 °C), at the input chambers [for example, the chambers x and y in Fig. 1(a)].

As the uncolored solution of the potassium iodide diffuses into the palladium chloride gel (yellow) it reacts with the palladium chloride to form iodo-palladium species. Sites where reaction occurs change their coloration from yellow

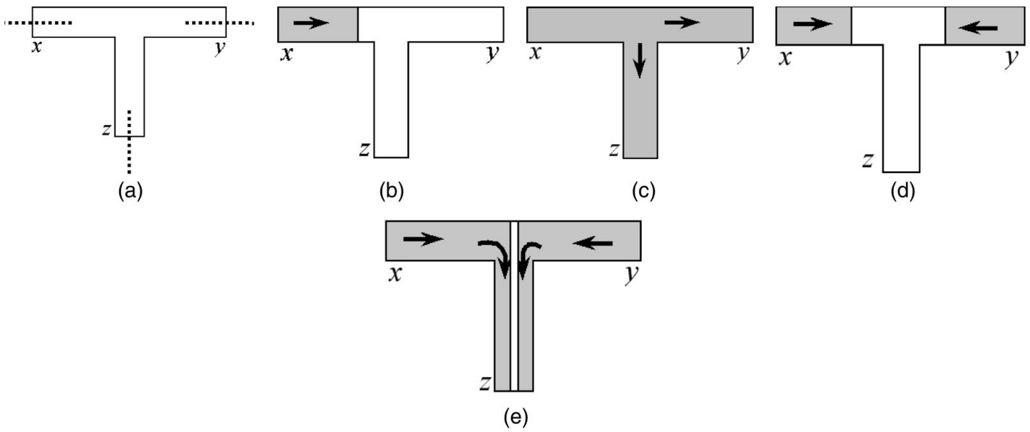


FIG. 1. Schematic action of the XOR gate. (a) An architecture of the gate; sites where state of the reactor is measured are shown by dotted lines. (b) One of the inputs gets a reactant, $x = T$, the reactant diffuses along the gate chambers and (c) reaches an output, $z = T$. (d) Both outputs get the reactant, $x = y = T$, two wave fronts of the reactant move towards one another, they interact and form an uncolored bisector (e), $z = F$.

(at a $PdCl_2$ concentration of 0.2%) to a dark brown (iodo-palladium species formed). It is assumed that the gel film is homogeneous and uniform and thus wave fronts originating from separate sources travel with the same speed and therefore the same distance to the point of collision. Sites of the substrate where two or more diffusive wave fronts meet (collide) remain uncolored; a bisector is formed separating the two (in this case) reactant sources. A more detailed treatment of the chemistry of this and other similar systems is available in previous publications [2,10,24,25].

There may be several reasons for the observed interactions of the wave fronts. It is likely that the mechanism is based on substrate competition between the advancing reactant wave fronts. As the diffusive wave fronts advance the speed of diffusion decreases. The fronts possess a chemical affinity for the substrate that is maximal at the initiation point of the reaction. This affinity decreases as diffusion speed decreases and the conversion of reactant + substrate to final product in the post front reactive zone increases. However, the physical presence of the diffusing wave front also exerts an influence over the gel directly in advance of it. A combination of both chemical and physical mechanisms causes substrate to move from the gel towards the diffusing front. Where the speed of diffusion is high this effect is minimal, however, when the speed of diffusion drops appreciably a dynamic concentration gradient of the substrate in advance of the wave front is formed. The upshot of this is that palladium chloride would be leached from the gel a small (but increasing proportional to reaction time—and therefore distance separating reactant sources) distance in front of a travelling wave front. Therefore, the unreacted KI eventually diffuses into a zone of the agar gel near the collision point that is depleted in the substrate resulting in the formation of substantially less PdI_2; the gel remains uncolored. Thus the bisector forms a permanent record of the substrates concentration gradient just prior to the collision of the fronts. Evidence for this mechanism comes from the bisector width which is proportional to the distance separating the reactant sources. If the sources are very close—no bisector or a diffuse bisector is formed if the sources are close (a few mm apart) then sharp thin bisectors are formed and if the sources are an appreciable distance apart wide more diffuse bisectors are formed (with an obvious concentration gradient).

III. XOR GATE

A *T*-shaped system of chambers comprises the gate: two horizontal chambers, *x* and *y* chambers, act as inputs and a vertical chamber, *z* chamber acts as an output. We measure the color of the input and output chambers in the sites shown in Fig. 1(a). When we apply a drop of reactant at one of the horizontal chambers, e.g., the *x* chamber, the reactant diffuses and spans along two other chambers, *y* and *z* chambers. All chambers become filled with precipitate and thus colored [Figs. 1(b) and (c)]. If two input chambers, are initiated via drops of the reactant [Fig. 1(d)] then the advancing wave fronts of the diffusing reactant interact at the intersection of the horizontal junctions and the vertical junction [Fig. 1(e)]. Thus an uncolored strip is formed between the two horizontal input chambers; this extends along the vertical section of the gate and corresponds to a ‘‘no precipitate’’ value at the measurement point of the output *z* chamber. The following mapping is implemented in the gate (*P* stands for precipitate, colored site and *U* stands for no precipitate, uncolored sites):

x chamber	*y* chamber	*z* chamber
U	*U*	*U*
U	*P*	*P*
P	*U*	*P*
P	*P*	*U*

Assume a colored (precipitate) site represents *T* (TRUE) and an uncolored site (no precipitate) *F* (FALSE) logical values, then we find that the gate realizes the function XOR: x XOR $y = (\neg x \wedge y) \vee (x \wedge \neg y)$. A negation is trivially realized as **T** XOR $x = \neg x$, where **T** is a constant truth.

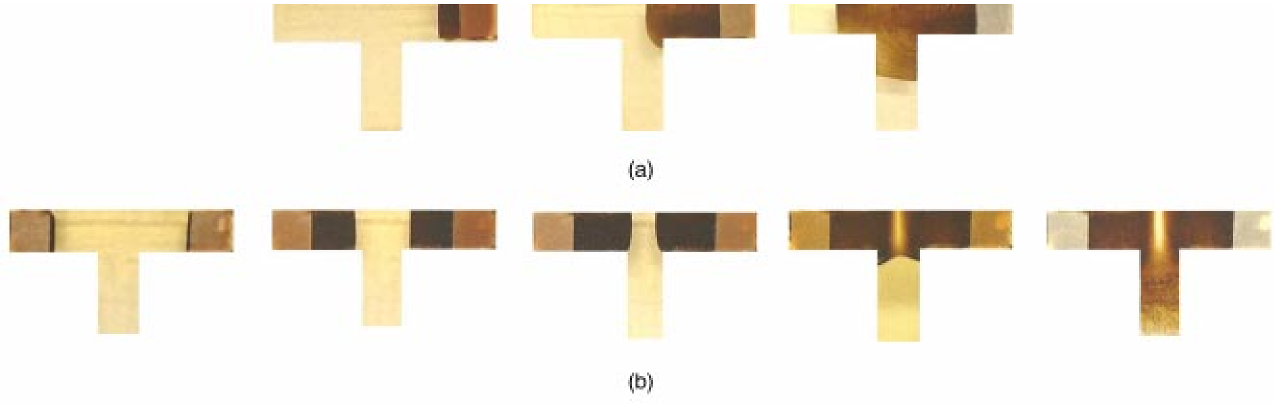
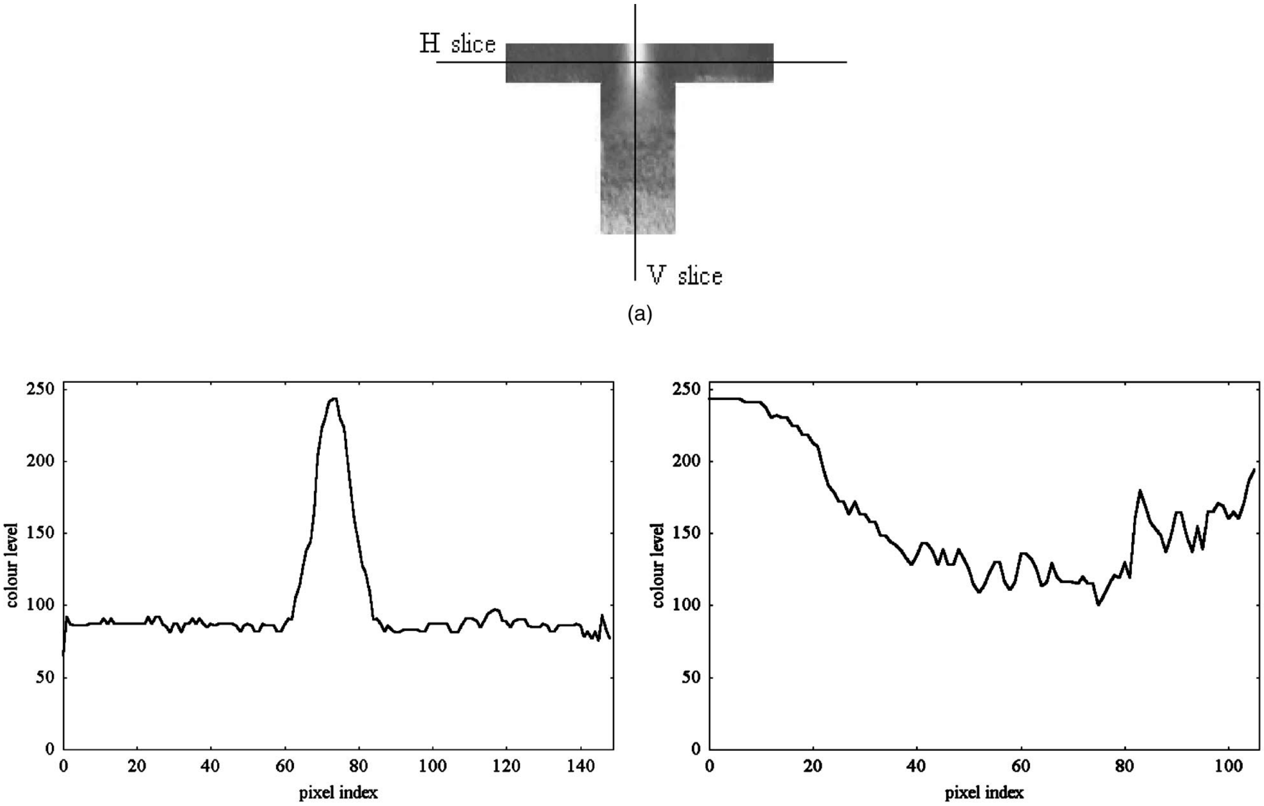


FIG. 2. Photographs of experimental implementation of the XOR gate. (a) The progression of the reaction-diffusion gate for the inputs $x=F$ and $y=T$. (b) The progression of the reaction-diffusion gate for the inputs $x=T$ and $y=T$.

Experiments with real chemical gates based on the reaction of a palladium chloride gel and a potassium iodide reactant solution are shown in Fig. 2. In this experimental gate the color intensity at given points gives a measure of the output variables as it directly correlates with the presence or absence, or relative concentration of the primary product (this is shown in details in Fig. 3). It is apparent that the experimental findings are slightly different from the theory although it can be seen that the XOR gate is implemented using this experimental setup. In the chemical system a distinct bisector is formed between the two directly opposed fronts emanating from the horizontal chambers *x* and *y*, however, this bisector does not extend far down the vertical output chamber *z*, thus a change in the measurement point would yield a more effective implementation of the XOR gate. Instead as the wave fronts diffuse away from the collision point and into the vertical output chamber the bisector becomes increasingly diffuse. However, it is obvious that there is a marked concentration difference at the output chamber corresponding to the interaction of the wave fronts.



(b) H slice

(c) H slice

FIG. 3. An example of the value T representation in concentration of precipitate. The diagram shows a one-pixel horizontal (b) and vertical (c), see scheme (a), slices of the 8-bit-per-pixel image of the experimental realization of the XOR gate [Fig. 2(b), bottom image].

FIG. 4. Implementation of AND gate in an unstable chemical reaction-diffusion system: the reaction of copper chloride and potassium hydroxide. The dark coloration is due to the secondary formation of copper oxide which occurs when the reaction becomes constrained behind a solid boundary. (a) The gate in the input state $x=F$ and $y=T$, reactant only at one input chamber. (b) The gate in $x=T$ and $y=T$ input state, reactant is presented in both input chambers. The increased stability of the system resulting in precipitate at the vertical output channel is apparent.

Thus the output variable of F is achieved in this experimental setup.

As the mechanism of bisector formation is based on substrate competition between the advancing fronts it can be seen how refining the experimental setup would yield a more effective implementation of the XOR gate. If the initial conditions were devised whereby the output chamber would be completely depleted of substrate at the point of collision between the fronts then the reaction could not proceed further and the fronts are effectively annihilated—the output chamber contains no precipitate. As discussed in Sec. II, a special case exists where two reactant sources are separated by an extremely small distance then no bisector is formed. This affords the possibility of constructing an AND gate from an adjusted experimental setup, where a length just below this critical distance separates the two input variables.

The palladium chloride gel—potassium iodide reaction belongs to a class of reactions that can be utilized in the same way to construct XOR gates. These include the reaction of a potassium ferrocyanide gels with a range of transition metal salts [25] and the reaction of copper (II) salts with sodium hydroxide [26]. It is probable that under the right conditions and carried out in geometrically constrained media that this implementation would extend to a wide range of chemical reactions of this type. However, some of the systems mentioned above have important differences when compared to the palladium chloride system. At certain substrate/reactant concentrations (specific to each reaction) the diffusive fronts are subject to spontaneous splitting caused by a chemical instability and the fronts cease to progress. Therefore, it is apparent that changing either concentration in the right range will result in a negation (any input results in an output of F) rather than an XOR gate.

IV. TOWARDS AND GATE

The mechanism of substrate competition between the fronts may allow an unstable negation gate to be converted into a true AND gate. In an unstable system any input variable results in an output of F however, if conditions are chosen carefully the stability of two otherwise unstable fronts may be maintained only when there is an input variable (reactant solution) at each point. Thus only when x and y chambers are initiated will any precipitate be detected at z. This is because the instability has its basis in exceeding a critical reagent concentration and only where two fronts compete for this same reagent will this concentration not be exceeded. Work to control nonlinear reactions of this type is complex practically although theoretically it is possible. Some preliminary results (Fig. 4) show a gate of this type [based on the reaction of copper (II) chloride (60 mg/ml) gel and potassium hydroxide ($8M$)] working in some capacity. The single input variable at y does not maintain stability and does not even reach the intersection point and the opening of the output chamber. The input variables at both x and y maintain stability until near the point of contact and subsequently precipitation can be seen to extend into the vertical output chamber z. The reproducibility of this gate implementation is low and it is included merely to serve as an indicator of future areas for development in using this class of nonlinear reactions for universal computation.

V. THREE-VALUED COMPOSITIONS

If we arrange measurement sites across the chambers [Fig. 5(a)] then an uncolored strip, call it S, can be detected. In the preceding section we constructed a binary composition, which happens to represent a logical XOR gate. What will be the structure of the algebraic system $A=\langle U,P,S,\bigcirc\rangle$? We already know the partial structure of the composition (we can put S instead of U as a result of the composition $P\bigcirc P$ because now we can sense a difference between uncolored chamber and an uncolored strip; symbols ? show not yet defined compositions):

$\bigcirc$	U	P	S
U	U	P	?
P	P	S	?
S	?	?	?

Let us reconstruct the rest of the table. How does an uncolored site U interact with the strip S? It is not sensible to generate a strip *per se* so we consider the state S as an input

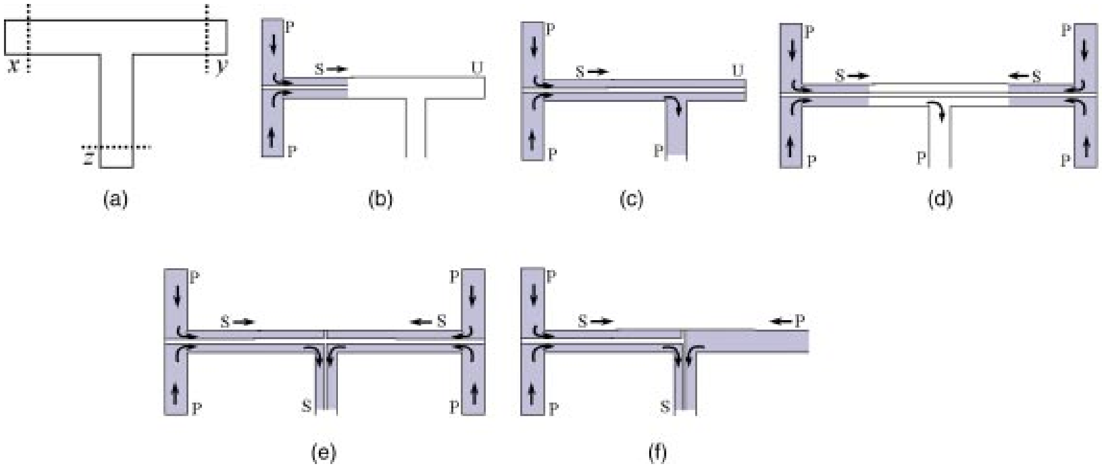


FIG. 5. Three-valued gate. (a) Dotted lines show sites where measurements are taken. (b)–(c) Two snapshots of the implementation of $S\bigcirc U=P$. (d), (e) Implementation of the composition $S\bigcirc S=S$. (f) Final configuration of a gate which performed computation $S\bigcirc P=S$.

to the gate from another gate [Figs. 5(b) and 5(c)]. First, the area with uncolored strip is generated via collision of two diffusive wave fronts. The strip moves along the horizontal chamber as shown in Fig. 5(b). When the growing structure gets to the branching point the upper part continues to spread, while the bottom part releases a reactant to diffuse to the vertical (output) branch, which becomes colored [Fig. 5(c)]. Thus we implemented the composition $S\bigcirc U=U\bigcirc S=P$. Consider two uncolored strips that move towards one another [Fig. 5(d)], then the upper wave fronts interact and form a short segment of uncolored sites and a long vertical segment of uncolored sites [Fig. 5(e)], thus we have implementation of $S\bigcirc S=S$.

The composition $S\bigcirc P=P\bigcirc S=S$ is realized in a similar manner [Fig. 5(f)]. So, the completed table looks as follows:

$\bigcirc$	U	P	S
U	U	P	P
P	P	S	S
S	P	S	S

The table gives the structure of the algebra A. The algebra A is commutative but not associative [e.g., $S\bigcirc(P\bigcirc U)=S$ and $(S\bigcirc P)\bigcirc U=P$]. It does not have minimal generators, neither units nor zeros. There are just two idempotents, U and S.

We are unaware of any published logics that might have the composition $\bigcirc$ as one of its operators. To construct a three-valued logical system we must represent logical values by the states U, P, and S. Three assignments of logical values may look sensible (however not truly satisfactory), * is a third logical value, usually interpreted as nonsense:

$E_1(S=T,P=F,U=*)$

$\bigcirc$	T	F	$*$
T	T	T	F
F	T	T	F
$*$	F	F	$*$

$E_2(S=T,U=F,P=*)$

$\bigcirc$	T	F	$*$
T	T	$*$	T
F	$*$	F	$*$
$*$	T	$*$	T

$E_3(U=T,P=F,S=*)$

$\bigcirc$	T	F	$*$
T	T	F	F
F	F	$*$	$*$
$*$	F	$*$	$*$

An operator represented by the table E_1 gives a constant T at the set $\{T,F\}$. The tables E_2 and E_3 could, in principle, be considered as tables of conjunction operators $\wedge_2$ and $\wedge_3$. The composition $*\wedge_2^*=T$ makes the gate $\wedge_2$ useless. The $\wedge_3$ gate may be applied for certain types of nonclassical reasoning if proper interpretation to $F\wedge_3 F=*$ and $*\wedge_3^*=T$ will be found. We leave this for future studies.

A chemical implementation of a three-valued composition is shown in Fig. 6 and equates to the composition shown in Fig. 5(e). The gates must be implemented on a much smaller scale to allow time for completion of the reaction before syneresis of the gel affects the results. Clearly the fact that the uncolored strip does not extend to the output variable in this form gives some differences between experimental and theoretical results. The real systems prove to have an in built smoothing mechanism. However, this said the output from each single gate corresponds directly to the concentration of products—which is given by the relative composition of reactants. Therefore, the output from each type of gate is distinct and its exerted influence on the next gate must be distinct depending on the configuration of those gates. For example in Fig. 5(e) the reactant concentration at the input of

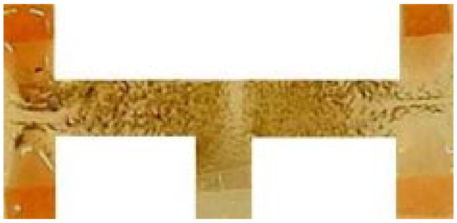


FIG. 6. A chemical implementation of a three-valued logic $S \bigcirc S = S$ gate [see scheme in Figs. 5(d) and 5(e)] in palladium chloride reaction-diffusion processor. The analysis of the results is complicated but the main point is that it shows combinations of gates can be achieved in these real chemical systems. Some leaching of the product is apparent in the original gates due to the high relative concentrations of the reactant—however a bisector (*S*) is still apparent in these original gates. A secondary bisector (*S*) can also be observed in the third gate—the width and intensity of this bisector should give a measure of the original input states of gates the first and the second gates.

the second gate will be double what it would have been if single input gates were utilized in the first stage (so effectively width of uncolored strip S equals half a width of precipitate representation of P). Therefore, in this case the strip S formed at the second junction will be approximately twice as wide as if single input variable gates formed the inputs to this gate.

Eventually reactant depletion will control the output variables from these combined systems—and could be used to differentiate between distinct input histories. For example the combination of various two input gates will eventually produce a precipitate (output—albeit mediated by collisions) where single input gates have become totally depleted in substrate and thus produce no output.

VI. CONCLUSION

The paper uncovers findings in structured computing in reaction-diffusion media, where the results of computation are given as a stationary spatial distribution of precipitate concentration. The chemical processor, described in the paper, is a disposable device, it could not be re-used as is the case with gates operating in a conventional computer. This is a disadvantage of the palladium processor, particularly when we compare it with excitable chemical media. However, the stationary spatial configuration of the processor could be interpreted as evolved hardware [27]; so, our results could be the first step in designing evolving chemical processors.

We discussed a laboratory prototype of a reaction-diffusion XOR gate. The XOR gate is employed in almost any field of computer science: from real life circuits, self-testing systems, error correcting codes, cryptographic systems to image processing and arithmetic logic unit design to biologically realistic algorithms of learning. So, ideally, our design could be applied in fabrication of all-wet unconventional computers and reaction-diffusion information processing units.

Also the paper proved, from common sense point of view, that reaction-diffusion chemical systems are general purpose computers—the palladium processor, which does logical computation in the paper, was previously applied to some problems of computational geometry, image processing, and path planning [2,10,25].

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