

# Nonlinear Chemical Dynamics and Its Interdisciplinary Impact: Dedicated to Ken Showalter on the Occasion of his 70th Birthday

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## INTRODUCTION

In Chemistry, the study of nonlinear phenomena and dynamic systems has its historic roots in seemingly unconnected experimental observations such as electrochemical oscillations,<sup>1</sup> periodic precipitation structures,<sup>2</sup> and propagating chemical waves.<sup>3,4</sup> For liquid-phase reactions, the latter were first described by Luther in 1906.<sup>4</sup> However, it was primarily the discovery and—perhaps more importantly—the early systematic exploration of the Belousov-Zhabotinsky (BZ) reaction<sup>5,6</sup> that combined with theoretical work by Prigogine and co-workers<sup>7,8</sup> nucleated the research field of nonlinear chemical dynamics. One of the pioneers, who subsequently developed this exciting field, is Kenneth Showalter who we honor with this festschrift on the occasion of his 70th birthday.

The earliest work of Ken Showalter, back then with Richard Noyes at the University of Oregon, focused on limit-cycle behavior and excitation waves in the BZ reaction. Subsequently in 1978, Ken joined the faculty at West Virginia University and began the vigorous investigation of chemical reaction mechanisms behind nonlinear phenomena such as bistability, oscillations, reaction-diffusion waves, period-doubling, and chaos. In 1989, he was named the Eberly Distinguished Professor of Chemistry. During the early 1990s, his group discovered rotating spiral waves on spherical surfaces<sup>9</sup> and explored front-instabilities,<sup>10</sup> chemical wave logic gates,<sup>11</sup> as well as shortest-path finding.<sup>12</sup> He also pioneered new methods for controlling low-dimensional chaos with specific emphasis on applications in chemistry.<sup>13</sup> In 1996, Ken was awarded the C.

Eugene Bennett Chair in Chemistry. Later work included studies of spatio-temporal chaos,<sup>14,15</sup> noise-sustained wave patterns,<sup>16</sup> and feedback control of waves.<sup>17</sup> During the past decade, his interests include synchronization phenomena among chemical oscillators, quorum sensing,<sup>18</sup> phase-clusters,<sup>19</sup> and chimeras.<sup>19,20</sup> During this 45-year journey through a scientific wonderland of nonlinearities and self-organization, Ken has led the way in demonstrating the relevance of new theoretical concepts in chemistry and in doing so also advanced the wider theoretical progress. Several of his students and postdocs hold faculty positions across the globe today, and many others have benefited from his cofounding (together with Harry Swinney and Raj Roy) of the “Hands-On Research in Complex Systems Schools,” which introduce students from developing countries to table-top scientific research on problems at the frontiers of science. Throughout his career, Ken received prestigious awards that include the Alexander von Humboldt Senior Scientist Award, the Bourke Lectureship from the Royal Society of Chemistry, and the Distinguished Alumnus Award from Fort Lewis College. His scientific reputation is obvious from his serving on a series of editorial boards, including *Chaos*, *Journal of Physical Chemistry*, *International Journal of Bifurcation and Chaos*, and *Faraday Transactions*.

In this Focus Issue, we explore some of the fruits of Ken’s contributions to nonlinear chemical dynamics by presenting a kaleidoscope of contemporary research illustrating the impact of nonlinear chemical dynamics on other disciplines ranging from theoretical physics and materials science to biology. To some extent,

this interdisciplinary impact has been more profound than the field's influence on chemistry itself. Nonetheless, there are also new research directions in chemistry, which in many ways are closely related to nonlinear dynamics and dissipative structures. Prominent examples include systems chemistry,<sup>21</sup> origins-of-life research,<sup>22</sup> chemobionics,<sup>23</sup> and work on adaptive and active materials.<sup>24</sup>

The interdisciplinary motivation for and the impact of nonlinear chemical dynamics are apparent in many works.<sup>25</sup> Returning to the oldest roots of the field, we see that Ralph Lillie's work on wave propagation on passivated steel was motivated by interest in "the essential factor in the transmission of the state of excitation from region to region in an irritable protoplasmic system (such as a nerve)."<sup>1</sup> In 1896, Raphael Liesegang wrote "Nearly all studies aimed at the elucidation of the mysterious processes in living beings were studies on the living organisms themselves. Only few modern researchers have tried, like the alchemists, to imitate these phenomena of life with nonliving matter."<sup>2,26</sup> Today, this visionary and long ignored statement could serve as the rallying call for various efforts in 21st century materials science and engineering. Links to biology and biomedicine are also apparent in the 1946 work of Wiener and Rosenblueth on spiral waves in cellular automata.<sup>27</sup> This first work on two-dimensional excitation waves demonstrated the possibility of vortexlike wave rotation around unexcitable obstacles and discussed the relevance to certain cardiac arrhythmias. In recent decades, the study of pinned chemical spirals and scroll waves has attracted much attention, and this work continues to draw its motivation in large parts from its relevance to cardiology.<sup>28</sup>

## IN THIS ISSUE

The individual articles in this Focus Issue can be loosely sorted into applications of nonlinear dynamics to chemistry, materials science, and biology. The chemistry-centered contributions include studies in the subareas of electrochemistry and chemohydrodynamics, of which the latter one has overlap with fluid dynamics and chemical engineering. In the following, we summarize these papers and show commonalities while also emphasizing connections to work by Ken Showalter.

## Chemistry

On the more chemistry-centered end of this Focus Issue, we find the article by Horvath *et al.*<sup>29</sup> who analyze coupled chemical oscillators via numerical simulations of a chemically realistic model of the BZ reaction. The investigation of such systems has a long tradition in chemical nonlinear dynamics and dates back to studies of coupled, continuously-stirred tank reactors from the late 1980s. More recently, these efforts retook the limelight with the growing interest in newly discovered phenomena such as phase clusters and chimeras. Horvath *et al.* focus on the elementary example of a pair of oscillators and discuss both, excitatory or inhibitory perturbations. They introduce a new phase-frequency model, which allows the reproduction and prediction of the behavior exhibited by pulse-coupled oscillators even though they extracted model parameters from single oscillator experiments. This tool can be extended to large-scale systems without in-depth knowledge of the underlying chemistry.

The external forcing of a single oscillator is discussed by Kumar *et al.*<sup>30</sup> for the interesting example of the "beating mercury heart," which refers to chemomechanical oscillations of a mercury drop in an electrolyte containing diluted sulfuric acid and dichromate ions. These oscillations arise from electrocapillarity (the dependence of the surface tension on the electrochemical potential) and have been known since the 19th century.<sup>31</sup> The authors report entrainment effects and find several Arnold tongues. While these are well-known phenomena for other chemical oscillators, this system also involves characteristic shape changes of the mercury drop. For example, 1:1 entrainment coincides with circular and elliptical modes, whereas 1:2 entrainment can result in beating triangular shapes.

An electrochemical phenomenon is also the focus in the work by Tosolini *et al.*<sup>32</sup> who investigated the electrodisolution of p-type silicon. The authors describe birhythmicity in this system, which indicates the existence of at least two distinct feedback loops. The dissolution process gives rise to chaotic behavior that develops via a period-doubling cascade and a quasiperiodic route with a torus-breakdown. This finding establishes the system as one of the few experiments with bichaoticity.

Electrodisolution is further explored in the paper by Hankins *et al.*<sup>33</sup> but now in the context of the abrupt and gradual onset of synchronization due to dynamical quorum sensing. The oscillators in this study are single-cathode multianode systems in which the nickel-based electrodes very slowly dissolve. Hankins *et al.* specifically show that this arrangement creates electrical interactions that are formally similar to quorum sensing and allow to "sense" the number of anodes or their effective "population density." Both the studies of Tosolini *et al.*<sup>32</sup> and Hankins *et al.*<sup>33</sup> extend the work of Ken Showalter on chemical chaos, chaos control, and synchronization phenomena. For example, his team was the first to demonstrate chaos control in the BZ reaction using a map-based, proportional feedback algorithm that stabilized periodic behavior.<sup>13</sup> His team also carefully investigated the synchronization of catalyst-loaded microbeads immersed in a catalyst-free BZ solution<sup>18</sup> and the emergence of chimera and phase-cluster states.<sup>19</sup>

Another fascinating—but for some experimental studies also highly undesired—aspect of chemical waves is the generation of fluid flow. The coupling between chemical reactions, diffusion, and fluid dynamics can arise through different effects such as surface-tension-induced or density-driven flows. Work in this wider field has seen tremendous progress since the late 1980s and early 1990s when many colleagues deemed the study of reaction-diffusion-convection phenomena to be too complicated. Today, we better understand its importance to pressing societal and environmental phenomena ranging from algae blooms<sup>34</sup> to CO<sub>2</sub> sequestration in saline aquifers or aging oil fields.<sup>35</sup> In our focus issue, this research thrust is represented by studies on reaction-driven oscillating viscous fingering<sup>36</sup> and simultaneous fingering, double-diffusive convection and thermal plumes.<sup>37</sup> Specifically, Rana and De Wit<sup>36</sup> report numerical simulations of a reaction-diffusion-convection model that reveals an active coupling between oscillatory chemical kinetics and the viscously-driven instability. Surprisingly, the oscillating kinetics can trigger viscous fingering for initially viscously stable situations, and changes in the viscosity profile can induce oscillations in an initially nonoscillating medium. A part of this study involves the famous Brusselator model that was introduced by Ilya Prigogine and René Lefever at the

Université libre de Bruxelles,<sup>38</sup> where Ken Showalter spent a sabbatical in the mid-1990s.<sup>39</sup> The second chemohydrodynamic study in this festschrift is also linked to Ken as his author, Reuben Simoyi, is a former, long-time faculty colleague. Simoyi<sup>37</sup> describes experiments in Hele-Shaw cells, involving the exothermic autocatalytic reaction of chlorite and thiourea. Observations include plumes and fingering patterns, which alternate as the main reaction front propagates through the reaction medium.

## Materials science

Applications of nonlinear chemical dynamics to materials science are a fascinating but still widely unexplored research direction. While many of the classic model systems either are homogeneous, liquid phase systems (e.g., the BZ reaction) or react gases via heterogeneous catalysis<sup>40</sup> (e.g., the oxidation of CO on Pt surfaces), only few examples result in permanent products such as plastics, gels, amorphous solids, or crystals. Notable examples are the (i) aforementioned Liesegang patterns,<sup>2,41</sup> (ii) inorganic, polycrystalline microstructures called “biomorphs,”<sup>41,42</sup> (iii) precipitation reactions forming tubelike membranes known as chemical gardens,<sup>23</sup> and (iv) other precipitation and crystallization phenomena.<sup>43,44</sup> For applications in materials science and engineering, the formation of a solid product is obviously only one necessary criterion. Another essential factor is to achieve tight control over the forming materials and shapes. The feasibility of establishing such control is clearly demonstrated by living systems that form a multitude of complex solid structures such as silica frustules, skeletons of glass sponges, nacre, tooth enamel, and even more complicated materials such as bones.<sup>45</sup> It is hence not surprising that the US-American Basic Energy Sciences Advisory Committee identified the following vision in their Grand Challenges report: “How do we characterize and control matter away—especially far away—from equilibrium?”<sup>46</sup> Clearly, the work of Ken Showalter provided much of the intellectual foundation for tackling this grand challenge. He introduced and demonstrated innovative methodologies for such control. Beyond his work on controlling chaos, we emphasize his pioneering contributions to the feedback stabilization of unstable propagating chemical waves and his numerous other publications utilizing the photosensitivity of the Ru(pby)<sub>3</sub>-catalyzed BZ reaction, in which visible light allows for the local and temporary inhibition of wave propagation.<sup>16,20,47,48</sup>

In this Focus Issue, two articles discuss the interdisciplinary impact of nonlinear chemical dynamics in materials science. Using numerical simulations, Potari *et al.*<sup>49</sup> investigate a precipitation reaction that is driven by a gravity current when a denser fluid is injected into an initially stagnant liquid. The flow field generated around the advancing liquid creates three spatially distinct zones where different modes of transport processes dominate. Depending on the relation between the chemical and hydrodynamic time scales, the emergent precipitate pattern associated with each zone can lead to different microscopic structures due to the presence of various gradients that allow additional thermodynamic forces to act. Malchow *et al.*<sup>50</sup> investigate the growth of lifelike inorganic microstructures, which form when an aqueous solution containing barium and silicate ions reacts with atmospheric carbon dioxide. The shape repertoire of these “biomorphs” include helices, funnels, urns, and corals that are the focus of this study. The authors introduce a three-step reaction

model that in conjunction with diffusion successfully simulates the formation of the experimentally observed coral-like structures. The model is related to the Gray-Scott model,<sup>51</sup> which was first attracting interest due to the existence of isolas and multistability (also research topics of Ken Showalter<sup>52,53</sup>) and later due to spot splitting and front instabilities.<sup>54</sup> The contributions by Potari *et al.* and Malchow *et al.* continue, in many ways, the work of Ken and explore the possible use of front instabilities and reaction-induced convection for the shaping of materials at mesoscopic and macroscopic length scales. Obviously, these are only modest, early steps and the technological application of nonlinear chemical dynamics to materials science remains a widely uncharted but highly promising field of research.

## Biology and biomedicine

More established links and applications of nonlinear chemical dynamics exist in biology and biomedicine. Long recognized examples<sup>27,55</sup> include dynamic similarities between nonlinear reaction-diffusion waves and propagating action potentials in neuronal and cardiac tissue; the cable equation describes spatio-temporal changes in electric potential in terms of a second spatial and first time derivative akin to Fick's second law of diffusion. Neuronal networks go beyond a simple active medium and provide a rich ground for nonlinear dynamics. In this Focus Issue, three contributions investigate different facets of such networks. Kaminker and Wackerbauer<sup>56</sup> report on alternating activity patterns and a chimera-like state in a ring network of diffusively and globally coupled excitable Morris-Lecar neurons. The underlying basic model<sup>57</sup> was developed in the early 1980s to describe different oscillatory dynamics in the muscle fiber of the world's largest barnacle (*Balanus nubilus*) and explicitly considers Ca<sup>2+</sup> and K<sup>+</sup> conductance. The neuronal network of excitable Morris-Lecar neurons exhibits transient spatio-temporal chaos with a sudden, system-intrinsic collapse to the rest state, where all neurons are inactive or to propagating pulses of neuron activity. Adding symmetric, global synaptic coupling spontaneously separates the chaotic firing pattern into a domain of irregular neuron activity and a domain of inactive neurons. Santos *et al.*<sup>58</sup> study a network of adaptive exponential integrate-and-fire neurons connected by chemical synapses. They find the coexistence of coherent and decoherent domains in the network as well as in the multicenter chimera states. Bera *et al.*<sup>59</sup> study a neuronal network of coupled Hindmarsh-Rose oscillators<sup>60</sup> (originally conceived for the study of spiking-bursting behavior of membrane potentials) with synaptic coupling. The authors find a new type of “spike chimera” where a domain with desynchronized spikes exists together with a domain of coherent quiescent states. These spike chimeras alternate temporally with a fully coherent state where all neurons are quiescent. In addition, a broad subset of the parameter space features several other dynamical states coexisting with chimeras.

Glycolytic oscillations are modeled in the study by Amemiya *et al.*<sup>61</sup> with emphasis on heterogeneities in the frequently studied HeLa cervical cancer cells—an immortal cell line named after the cancer patient, Henrietta Lacks, who died in 1951. In response to starvation, these cells exhibit large variations in the period and the overall duration of oscillations. The authors find that this heterogeneity is caused by variations in the rate constants of enzymatic reactions. Specifically, the activity of the glucose transporter appears



to be important for determining whether oscillations appear, whereas the initial concentrations of metabolites have only small effects.

Lastly, we return to quorum sensing in the context of biochemical reactions. Markovic *et al.*<sup>62</sup> analyze enzyme-loaded agarose beads and the effect of chemohydrodynamics on their quorum sensing abilities. In this system, reaction-induced pH changes cause the formation of plumes that physically move the beads through the system. This study is complemented by numerical simulations and combines in many ways the different subtopics of this Focus Issue: nonlinear dynamics, specifically phenomena such as oscillations, traveling waves, quorum sensing and chimeras, spatial coupling via diffusion or similar processes, chemically-driven convection and particle motion, and last but not least, the inherent interdisciplinarity of all of these efforts.

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