

Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting
August 23-26 & August 30-September 1, 2021

Novel Methods for International Safeguard Sensing: Nonlinear Chemical Waves and In Situ Logic-Tree Analysis

Thomas Dewers

Nuclear Waste Disposal
Research & Analysis,
Sandia National
Laboratories, Albuquerque,
NM, USA

Jason Heath

Geomechanics, Sandia
National Laboratories,
Albuquerque, NM, USA

Kristopher Kuhlman

Applied Systems Analysis
& Research, Sandia
National Laboratories,
Albuquerque, NM, USA

Richard Jensen

Geomechanics, Sandia
National Laboratories,
Albuquerque, NM, USA

Jacob Harvey

Geochemistry, Sandia
National Laboratories,
Albuquerque, NM, USA

Robert Finch

Internal Safeguards &
Engagements, Sandia
National Laboratories,
Albuquerque, NM, USA

Abstract

Chemical waves are self-sustaining fluctuations in chemical concentrations that arise from nonlinear coupling of transport and chemical reaction in a fluid medium. Chemical waves respond to and retain a history of encountered stimuli, and the waves propagate spatially even in the absence of advection. We investigate use of gas-aerosol and aqueous phase chemical wave systems as a basis for novel methods of Safeguards sensing. Amplitude and frequency of chemical waves can be tuned in sensitivity to indicate presence of targeted products or signatures, and the waves can be triggered by the trace presence of solid, gaseous, or liquid chemical trace compounds, physico-acoustic perturbations, magnetic fields, and optical stimulation. We use computational fluid dynamics modeling to demonstrate scenarios of detection; these include using an ozone-aerosol autocatalysis in detection of breaches in dry cask storage of spent fuel, and intrusion detection in a subsurface geologic repository for nuclear waste storage. Fault tree or event tree analysis involves representation of events or features of system as an interconnected series of Boolean operations or logic gates. The logic tree allows assessment of the root cause of a particular event, or an analysis of the cascading events that arise from a given event. We create a chemical-based tree analysis within the system of interest itself, using chemical waves as the “telegraph line” to transmit information and molecular logic gates as the Boolean logic gates of a tree analysis.

Introduction

Waves are energy disturbances that propagate through a medium. As such, chemical waves are true spatio-temporal waves stemming from nonlinear coupling of transport and chemical reaction, phase transition, or electrical excitation under far-from-equilibrium conditions (Nicolis and Prigogine, 1977; Ortoleva, 1992). These need not involve transport of mass (but can move with advective fronts) but rather transport of chemical information such as pH, redox state, and/or concentrations. Much of fundamental understanding on chemical waves owes to the famous Belousov-Zhabotinsky (BZ) reaction (Zaikin and Zhabotinsky, 1970; Winfree, 1984; Field et al., 1972) involving a metal-

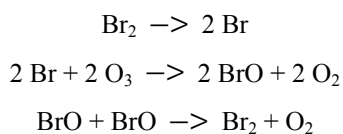
catalyzed oxidation of an organic substrate by bromate. The autocatalytic production of HBrO_2 , a positive feedback, gives rise to a range of processes including temporal oscillations between oxidized and reduced states, spatial propagation of chemical wave fronts of various morphologies (Mori et al., 1991), and bi-stability (i.e. steady state at “high” or “low” compositional states). Many workers have leveraged these behaviors into tools of “liquid chemical computing”: chemical waves interacting with a geometrical medium as chemical switches (Rossler, 1976); logic gates (Hjelmfelt and Ross, 1995); chemical neural networks and Turing machines (Hjelmfelt et al., 1991); and chemical clocks (Winfree, 2001). Generally, kinetic chemical systems with multiple stationary states coupled with forced advective transport can support chemical waves with the ability to provide information on spatial networks through which they propagate (Hjelmfelt et al., 1993; Steinbock et al., 1996).

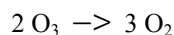
Chemical-wave based information processing has been invoked previously in image processing, robot navigation, and Boolean logic gates (Motoike and Adamatzky, 2004). Recent implementation has suggested processing down to the scale of a single protein molecule (Adamatzky, 2017). Originally proposed by Hjelmfelt et al., (1991), chemical logic gates have been constructed in analogy with neural networks that include AND, I AND NOT J, and NOR gates that form the basis of simple computers and even Turing machines. More advanced non-binary logical chemical “devices” have been proposed by Motoike and Adamatzky (2004). Generally, these concepts have advanced beyond the simple models by use of single capillary tubes (Kitahata et al., 2004), microfluidic cells (Ginn et al., 2004) or mesoporous media (Amemiya et al., 1995), and we have investigated this approach in a previous study (Jensen et al., 2018; Dewers et al., 2018, 2019a,b).

Herein we present the concept of chemical waves as geospatial sensors that respond to stimuli and transport information spatially. Through a series of mathematical models, we demonstrate liquid- and gas-phase oscillators that develop chemical waves. We show that the nature of chemical waves is sensitive to environmental conditions including geometry, radiation conditions, aerosol concentrations, pH, organic substrates, and other examples. We apply these concepts to Safeguards sensing in a subsurface nuclear waste repository; we also examine breaching of dry cask spent fuel storage and discuss the use of chemical wave sensing in other International Safeguards contexts. Contrasting with typical linear tracers, chemical waves can propagate faster than conservative tracers, can propagate against an advective gradient, and can nonlinearly amplify small perturbations.

Background: Chemical Waves as Sensors and Nonconventional Computing

Nonlinear chemical waves generally involve a degree of autocatalysis, whereby reaction products feedback into a reaction scheme as reactants. An important environmental example includes the destruction of ozone by chlorofluorohydrocarbons and other halogens (here we use bromide, Br) by an autocatalytic reaction step. In the reaction sequence below, ozone (O_3) is reduced into molecular oxygen (O_2) by the destruction and production of Br_2 . This is shown by summing the first three reactions to arrive at the fourth net reaction. To the extent that the reaction sequence reactants are also the products (and accompanied by the long residence time of Br and other halogens in the atmosphere) this reaction scheme is autocatalytic, i.e., the products are reactants.





The canonical liquid phase chemical oscillator is the Belousov Zhabotinsky reaction (Zaikin and Zhabotinsky, 1970; Field et al., 1972) which entails a metal-catalyzed oxidation of an organic substrate by bromate. We explore this reaction network at length, but the autocatalytic production of HBrO_2 is commonly visualized using an iron indicator. Figure 1 shows a picture of the bottom of a beaker involving this autocatalytic reaction network developing in spatially varying redox states, which constitute chemical waves in the present context.

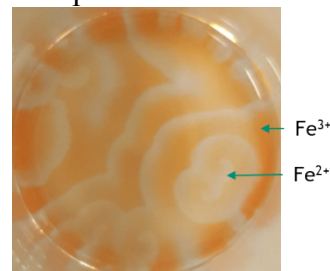


Figure 1. An example of the Belousov Zhabotinsky reaction involving Br-Fe oxidation and reductions. The different redox zones propagate through space, via a system of chemical waves. Note this involves spatial transport in redox state, but not necessarily mass (although mass does diffuse to a limited extent).

Methods

In what follows we examine (isothermal) autocatalytic reaction schemes in a variety of settings that we will utilize later in this paper. We do this by modeling the chemical kinetic equations in three contrasting modes (after simple chemical engineering design), shown in Figure 2.

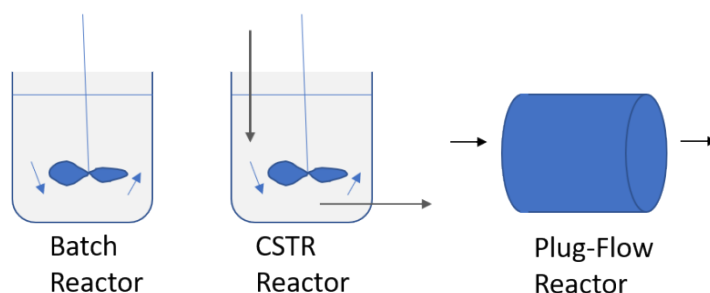
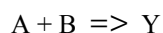


Figure 2. Three reactor designs useful for modeling autocatalytic reaction schemes and chemical waves. Batch reactors have a constant mass (closed system) and are well stirred, wiping out internal chemical gradients. CSTRs (continuously stirred tank reactors) have constant infeed and outfeed, and as such are open system reactors, but also are well stirred with no chemical gradients. Plug flow reactors have constant infeed and outfeed.

Batch reactors: Typical equations for reacting chemicals in batch mode in a single solvent medium involve so-called mass action kinetics and can include forward reactions, backward reactions, and/or reversible reactions. As an example, the reaction



involves an irreversible reaction with reactants A, B and product Y. A mass action description of the kinetics of this reaction would involve ordinary differential equations for A, B, and Y (brackets denote concentrations and first order mass action kinetics apply):

$$\frac{\partial[A]}{\partial t} = \frac{\partial[B]}{\partial t} = -\frac{\partial[Y]}{\partial t} = -k_f[A][B] = r$$

CSTRs: In the case of inlet and outlet flow rates equal to q , a simple model for the mass or mol balance of the component A above would be (for reactor volume V and reaction rate r_A equal to r above)

$$V \frac{\partial[A]}{\partial t} = [A]_{in}q - [A]q + Vr_A$$

In this equation, the reactor residence time would be V/q .

Reactive Transport Models: For a general reactive transport model, a set of mass and momentum conservation equations would include a Navier-Stokes equation (with velocity \mathbf{u} and pressure p)

$$\rho \frac{\partial \mathbf{u}}{\partial t} + \rho(\mathbf{u} \cdot \nabla)\mathbf{u} = \nabla \cdot [-p\mathbf{I} + \mathbf{K}]$$

coupled to a mass conservation relation (with a Boussinesq fluid of density r) and viscosity μ

$$\rho \nabla \cdot \mathbf{u} = 0, \quad \mathbf{K} = \mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)$$

These relations are coupled to a general reactive transport equation with concentrations c , diffusional/dispersive flux J , and overall reaction rate R

$$\frac{\partial c_i}{\partial t} + \nabla \cdot J_i + \mathbf{u} \cdot \nabla c_i = R_i$$

$$J_i = -D_i \nabla c_i, R_i = \vartheta_{ij} r \quad \text{and} \quad r_j = k_j^f \prod_i c_i^{-\vartheta_{ij}}$$

where D is a diffusion/dispersion coefficient, and the ϑ_{ij} are stoichiometric coefficients. The k_i are rate coefficients; this form assumes a mass-action kinetics, and in principle, other kinetic rate laws can be used, e.g., sorption reactions between fluid and solids, or between a gas phase and aerosols.

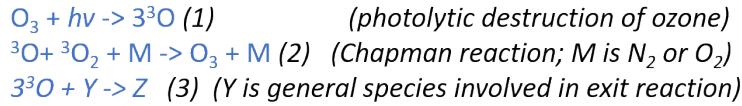
Results

Examples of Gas-Phase Oscillators

The previous autocatalytic reaction networks utilized aqueous phase reactions and generated chemical waves with wavelengths that are likely too small to be practical for larger-scale geospatial sensing (although logic gate applications for liquid phase computing based on the BZ schemes are common in the literature, e.g., Lavrova and Vanag, 2014; Adamasky, 2002; 2015; Steinbock et al., 1996; Hjelmfelt and Ross, 1995; Hjelmfelt et al., 1993). A relatively simple oscillator with larger wavelengths and operating in a gas phase was proposed by Jungwirth (2001) involving ozone.

This simple autocatalytic reaction scheme is photocatalytic, meaning it is driven by exposure to photons. Most generally for ozone described in terms of ultraviolet (UV) light sensitivity, a wide range of ionizing radiation wavelengths can influence the nature of the oscillator as we demonstrate here. In considering this system for geospatial sensing, some points to consider: coupling ozone with halogen-bearing aerosols changes wavelength and character of the limit cycle oscillations; potential for tunability of ozone sensing dynamics; chemical wave dynamics evident from spatio-temporal computational fluid dynamics (CFD) model on measurable time and length; brief induction time followed by chemical wave train that advances faster than advection; reaction is photoautocatalytic, which means that the oscillations are driven by exposure to light including UV and does not operate in the dark; and wavelength and amplitude of oscillations are sensitive to ionizing radiation, and as such are sensitive to radiation. This suggests an application in detecting radiation leaks. For example, ozone is decomposed orders of magnitude more rapidly by ionizing radiation compared to UV light (Harteck et al., 1965).

The autocatalytic nature of ozone decomposition was discussed by Jungwirth (2001) in the following simple reaction couple involving ozone O_3 , oxygen O_2 , and a metastable intermediate 3O :



The Chapman reaction is considered an important aspect of atmospheric ozone dynamics (Cao and Gutheil, 2011; Brauer et al., 2013). Three examples examining this oscillator driven by UV catalysis, radiolytic decomposition of ozone, and under dark conditions (with absence of oscillatory behavior) is shown in Figure 5. Different forms of radiation produce distinct variations in the frequency and magnitude of the oscillatory behavior, useful in sensing presence of radiation leaks.

Ozone couplings with halogens have been a topic of much interest since the 1970s and concerns of global ozone depletion. We show in Figure 4, using reaction rates and dynamics given in Herrmann et al. (2019), that simple coupling with Br aerosols has a tunable effect on ozone oscillatory behavior.

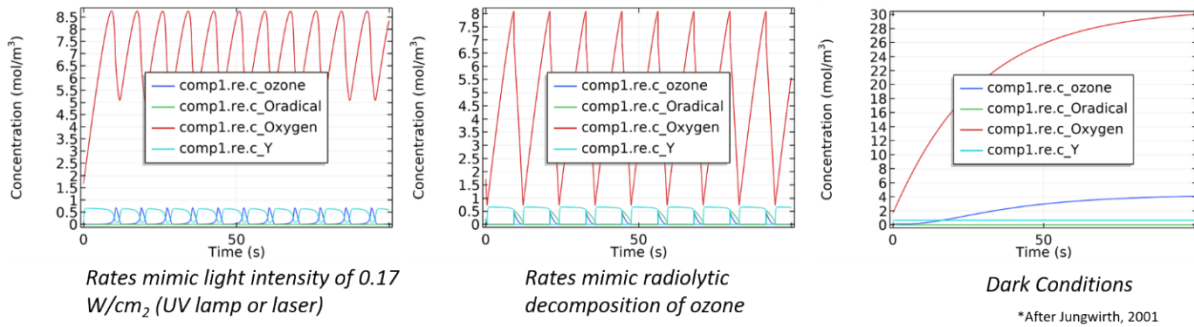


Figure 3. Examples of ozone catalysis by ionizing radiation and oscillatory behavior. The left panel shows UV-catalyzed ozone oscillations. The middle panel shows ozone decomposition by radiolysis (after Harteck et al., 1964), and the right panel shows how the system would operate in the dark, with no attending oscillatory behavior.

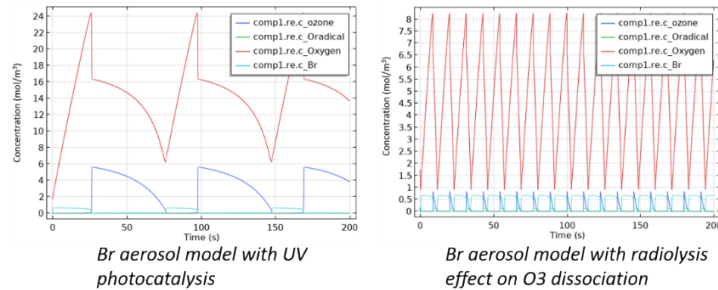


Figure 4. Ozone photocatalysis coupled with Br-aerosol dynamics, modeled after atmospheric O₃-Br reaction couplings of Brauer et al. (2013), Carpenter et al. (2013), and Evans et al. (2003). The left panel shows effects of the Br-aerosols on the oscillatory scheme in Figure 3, with a dramatic change in waveforms. The right panel shows the effects of Br aerosols coupled with radiolysis.

Safeguards Example 1: Dry Cask Storage Monitoring for Radiation Breaches

A three-dimensional example of using chemical wave sensing in detecting breaches in dry storage casks utilizes the simple gas-phase ozone oscillator described earlier and could be used outdoors in the presence of a steady wind advection. Such a scenario is portrayed in Figure 5, which shows variation in advective velocity among the simulated cylindrical dry storage casks.

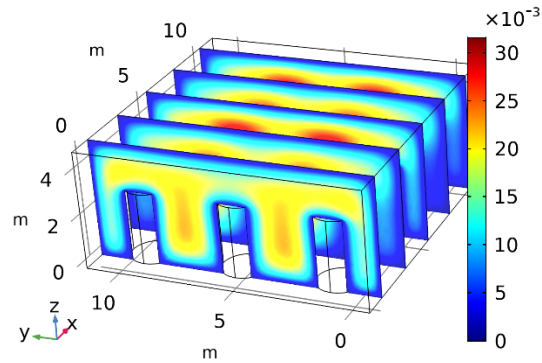


Figure 5. Variability in advective velocity, modeled using the CFD capability of COMSOL Multiphysics. The wind velocity at the domain boundary is $1.0E-3$ m/s and varies through and around the canisters.

Taking advantage of symmetries in the advective transport, we show in Figure 6 chemical wave development along with the advective transport, that passes from one end of the cask storage domain to the other in 900 seconds. In this case the ozone and associated gas-phase chemistry are released at the left-hand boundary at a constant rate developing wave trains that propagate across the domain.

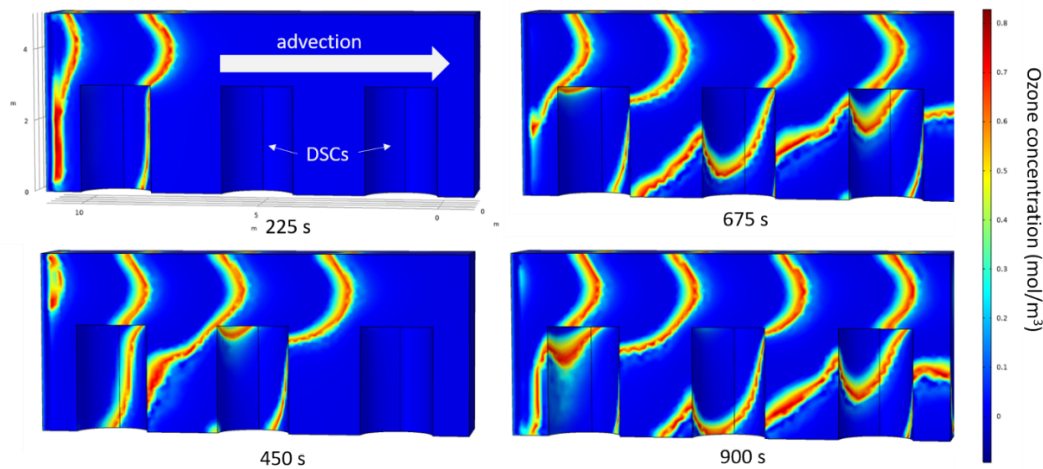


Figure 6. Ozone oscillator dynamics develop as chemical wave trains and propagate with an advective air front (wind).

In the case of zero wind (which also applies to enclosed buildings or galleries for geologic nuclear waste storage), Figure 7 shows an example where heightened point-source ozone destruction by radiolytic kinetics (as would attend a radiation leak) generates a localized chemical wave front. This simulation is initiated with the simulation domain filled with ozone and associated gas phase trace chemistry, and the ozone dynamics lead to batch-like fluctuations across the domain. However, after 1.5 s, a localized chemical wave is being generated at the bottom of the middle cask due to a point source, which simulates radiation leakage and ozone destruction via radiolysis. Thus, laser spectroscopic or other methods could detect localized radiation breaches in the canister overpacks.

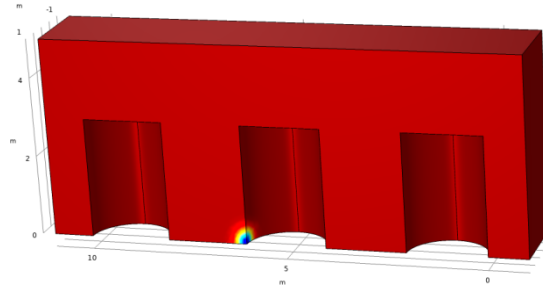


Figure 7. A localized radiation source in the lower left portion of the middle canister generates a localized chemical wave associated with radiolytic destruction of ozone. This demonstrates the sensing capability of nonlinear chemical wave sensors in *in materio* sensing of localized point source contamination.

Safeguards Example 2: Radionuclide Detection in Repository Disturbed Rock Zones

The previous gas-phase chemosensor systems could be applicable to above ground open-air dry cask storage systems, the interiors of buildings used for storage, or in underground geologic repositories in pre-closure monitoring scenarios. Another relevant scenario that we foresee for nonlinear chemosensors is assessing migration of radionuclides in aqueous systems as might occur in fractured rock or damaged zones around constructed subsurface engineered caverns. Here we outline how chemical wave sensing systems would behave in fractured media, and how such systems might be used for detection.

The Oregonator autocatalytic oscillator is often used to represent aqueous phase oscillatory systems (Field and Noyes, 1974) and was implemented in PFLOTRAN (www.pflotran.org), an open-source massively parallel reactive flow and porous media transport code. A heterogeneous two-dimensional system with binary properties (i.e., open fracture or impermeable rock) was used to represent a simplified system of fractures. Figure 8 shows the simplified fracture network used for the study. The arrangement two parallel groups of fractures resulted in a backbone of flowing fractures (red and pink in bottom panel) and a series of dead-end areas with minimal advective flow.

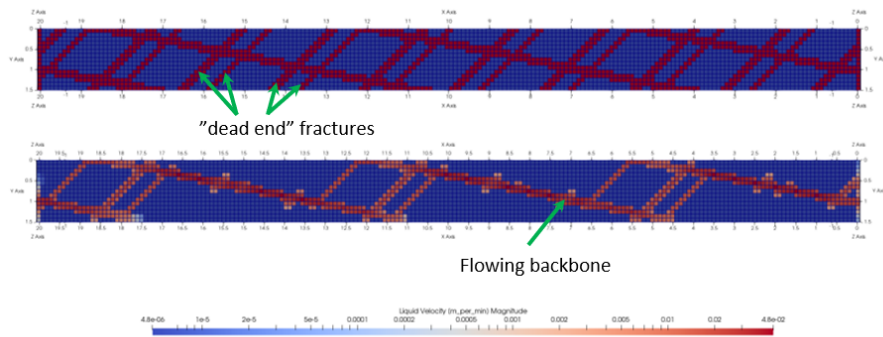


Figure 8. 2D Fracture PFLOTRAN model ($200 \times 15 \times 10$ cm square elements). Top panel shows fractures (red) and no-flow areas (blue). Bottom panel shows the steady-state flow velocity. Blue fractures in lower subplot are not flowing (i.e., dead ends); pink or red fractures are flowing.

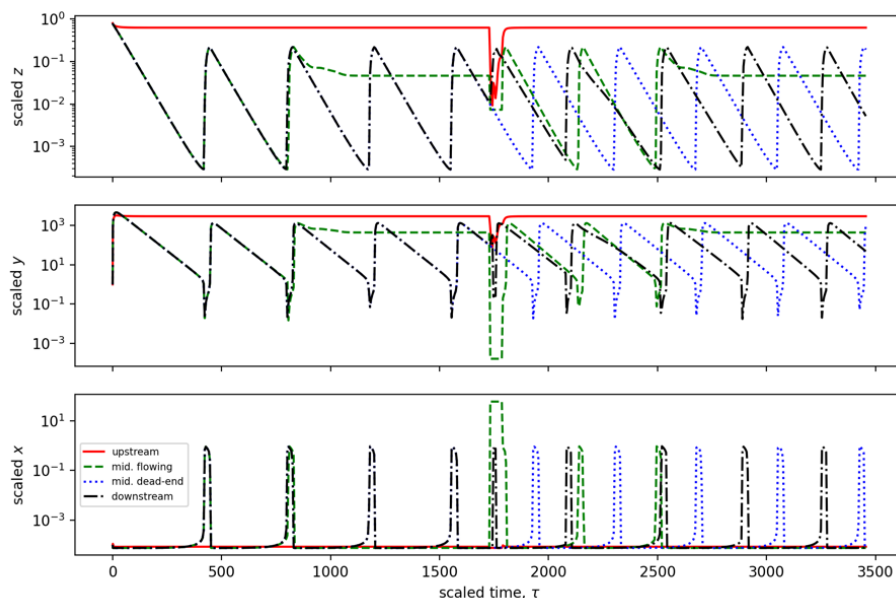


Figure 9. Scaled concentrations at observation locations in 2D Fracture PFLOTRAN model when a spike of X is introduced at $\tau = 1800$. Oscillations in the flowing fractures reset spatially.

When flowing water through the system, it was observed that there are two primary types of behaviors in the system: areas with significant advection, where oscillations stopped after a short time, and areas without significant advection, where oscillations continued to the end of the simulations. Comparing flowing fractures at the end of the simulation, we observe “standing waves” or oscillations-in-space have set up in the flowing fractures while oscillations in time continue in the non-flowing areas. We investigate the impact of a spike change in concentration into a flowing fracture near the mid-stream flowing fracture observation point. Figure shows scaled concentrations illustrating how the change in concentration resets the oscillations again (both in time and space), in a wave front moving away from the point of the disturbance.

The combination of the simplified Oregonator model with the more physically realistic 2D fracture network distribution allows exploration of the dynamics of this system more readily than more complex chemistry might allow. The system is stable and tends to an oscillation-in-time cycle at locations with minimal advection. At locations with significant flow, spatial waves set up, which can be disturbed by changes in concentration. Transient signals propagate away from the disturbance, setting up spatial waves of different character than those due to the original advection field. These stationary waves may leave a “fingerprint” behind for later observation from a transient disturbance that occurs too quickly or is too localized to be observed directly.

Discussion: Monitoring, Detection and Event Tree Decision-Making

Information read-out of the chemical waves is performed by measurement of the oscillating chemical concentrations, which may be performed at point locations or integrated along a measurement path, and which may be at different temporal resolutions, such as in real-time or by sampling with later laboratory analysis. Depending on the application, read-out may involve laser, absorption, and/or vibrational spectroscopy, or lab-based chromatography, spectroscopy, or mass spectrometry. For difficult-to-access host media such as a building with hazardous materials, laser-based spectroscopy with beam positioning by mirrors, beam-splitters, and detectors enables precision positioning for

measurements and ultimately defines the reachability region of measurements. The chemical waves themselves may propagate information out of regions that are otherwise inaccessible to where measurements could be made. Interpretations of the chemical waves is facilitated by understanding how and what triggers and controls the chemical wave patterns. We propose a logic-tree framework to map chemical responses to triggers and their cascading spatial patterns, which would be interpreted as a set of inter-related yes/no questions of a fault- or event-tree analysis. Thus, read-out would give the final results of the logic tree analysis that is performed *in materio* by the chemical waves themselves. Another interpretation approach is inverse modeling with coupled CFD and parameter estimation to infer the underlying triggers in the host media that produced chemical waves.

Applications for this chemical wave sensing-processing and data transmission system are diverse and include, but are not limited to, the following scenarios: identification of trace compounds in the host environment (e.g., on floors, walls, or the surfaces or insides of containers); seals or feature evaluation that allows determination if something has been disturbed, such as a container or door; identification of leaked substances may leak from a container or confinement region; counting of items by chemical wave products accumulating in proportion to interactions with the target item; connectivity assessment of tortuous pathways in a geometrically complex system; and event tree analysis through chemical wave logic circuits answering a linked set of yes/no questions, which may encompass many of the aforementioned scenarios. These scenarios can be embodied in a variety of industries, research settings, or similar avenues, such as: nuclear safeguards assessment such as those performed by the International Atomic Energy Agency for material accountancy, seal verification, surveillance, and intrusion detection in settings such as spent fuel storage in dry casks, spent fuel storage in pools, or underground nuclear waste disposal; inventory in large warehouses that involve geometrically complex packing with many nooks and crannies that are otherwise difficult to access; piping systems for chemical processing or reactors that may involve geometric complexity and features that need monitoring/sensing; subsurface systems that involve natural or induced fracturing such as enhanced geothermal systems where the connectivity is key to thermal profiles and performance.

Conclusions

- Through a series of increasingly complex simulation schemes for batch, CSTR, and spatiotemporal dependent chemical reactive transport, we demonstrate the use of nonlinear chemical waves as geospatial sensors.
- Chemical waves respond to and retain a history of encountered stimuli, and the waves propagate spatially even in the absence of advection. Chemical waves can be triggered by trace quantities of solid, gaseous, or liquid chemical compounds, physico-acoustic perturbations, radiation, magnetic fields, and optical/UV stimulation.
- The applied chemical compounds sense characteristics of a host medium (which could be gas or liquid phase) by producing diagnostic spatial and temporal patterns in chemical waves as triggered by what is in the host medium that is contacted by the chemicals. The chemical waves perform *in materio* analysis at different levels of complexity depending on the particular system and application, which may include: identification of particular substances or configurations of items in the host medium; verification that seals or other features have not been damaged or removed; and/or verification that containers have not leaked

We suggest that a class of pH oscillators (Luo and Epstein, 1991) may be useful in subsurface applications which could operate in existing or stimulated fracture networks; gas-phase or gas-

aerosol oscillators such as the demonstrated ozone systems herein could be useful for surface applications such as monitoring dry cask storage of spent fuel, or in detecting leakage in underground galleries for geologic storage of nuclear waste.

Acknowledgements

The authors thank the US DOE National Nuclear Security Administration, Office of Defense Nuclear Nonproliferation Research and Development, for funding this work. This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the US Department of Energy or the United States Government. SNL is managed and operated by NTESS under DOE NNSA contract DE-NA0003525.

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