Spatially organized partial synchronization through the chimera mechanism in a network of electrochemical reactions

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Detailed experimental and numerical results are presented about the pattern formation mechanism of spatially organized partially synchronized states in a networked chemical system with oscillatory metal dissolution. Numerical simulations of the reaction system are used to identify experimental conditions (heterogeneity, network topology, and coupling time-scale) under which the chemical reactions, which take place in a network, are split into coexisting coherent and incoherent domains through the chimera mechanism. Experiments are carried out with a network of twenty electrodes arranged in a ring with seven nearest neighbor couplings in both directions along the ring. The patterns are characterized by analyzing the oscillation frequencies and entrainments to the mean field of the phases of oscillations. The chimera state forms from two domains of elements: the chimera core in which the elements have identical frequencies and are entrained to their corresponding mean field and the chimera shell where the elements exhibit desynchrony with each other and the mean field. The experiments point out the importance of low level of heterogeneities (e.g., surface conditions) and optimal level of coupling strength and time-scale as necessary components for the realization of the chimera state. For systems with large heterogeneities, a ‘remnant’ chimera state is identified where the pattern is strongly affected by the presence of frequency clusters. The exploration of dynamical features with networked reactions could open up ways for identification of novel types of patterns that cannot be observed with reaction diffusion systems (with localized interactions) or with reactions under global constraints, coupling, or feedback.

1. Introduction

Activity patterns originating from competing chemical reactions in spatially extended systems are ubiquitous in nature and underlie important physiological processes, e.g., in circadian rhythms, neuron electrophysiology, or calcium signaling.\(^1\)\(^-\)\(^4\) The general features of patterns often can be explored with relatively simple chemical reactions;\(^2\)\(^,\)\(^3\) these studies facilitate the development of experimental and theoretical tools for effective characterization of complex, multiscale systems. One important class of patterns can arise in discrete reaction units with oscillatory chemical reactions. The units can interact with each other through mass transfer, electrical coupling, or some external constraint. Examples include coupled continuous, stirred tank reactor systems (CSTR) of Belousov–Zhabotinsky (BZ) oscillators,\(^5\)\(^-\)\(^8\) pH oscillators,\(^9\) or biochemical oscillators;\(^10\) large population of units can be obtained from BZ – beads,\(^11\)\(^,\)\(^12\) BZ – microwell arrays,\(^13\) or in BZ – micro\(^14\) and – nanoscale\(^15\) systems. In heterogeneous systems, electrochemical reactions on multielectrode arrays can be used for the investigation of oscillatory patterns.\(^16\)\(^-\)\(^21\)

A common spatiotemporal pattern observed with weakly, globally coupled oscillatory units is cluster formation;\(^22\) the population splits up into groups in which the kinetic trajectories are identical but differ from those of the other groups. One cluster states obtained from slightly heterogeneous oscillators (which have a frequency distribution) can develop through Kuramoto\(^19\)\(^,\)\(^23\) or quorum\(^15\) transitions. Two and higher order cluster states have been observed with electrochemical,\(^24\)\(^-\)\(^26\) BZ,\(^27\)\(^,\)\(^28\) and heterogeneous catalytic\(^29\) reaction systems. Another type of self-organized pattern consisting of co-existing phase coherent and phase incoherent elements, known as the chimera state, was found to exist in numerical simulations and theoretical studies of networked units.\(^17\)\(^,\)\(^27\)\(^,\)\(^30\)\(^-\)\(^37\) While with local and global interactions the typical behaviors are various types of reaction waves and clusters, respectively, with network coupling novel patterns are expected to arise because of the large variety of possible modes of spatiotemporal variations. Experiments in chemical BZ bead reaction,\(^27\)\(^,\)\(^35\) electrochemical corrosion\(^17\) and silicon dissolution reaction,\(^38\)\(^,\)\(^39\) mechanical pendulum system,\(^36\) and feedback modulated liquid crystal\(^37\) and optical\(^40\) systems have provided confirmation for the existence of the chimera state.
patterns. The dynamical varieties of these patterns imply that there are possibly many mechanisms that can generate chimera-like structures. A striking feature of the chimera state obtained from the original Kuramoto-mechanism34 is that both two locally coupled and a population of globally coupled identical oscillators form a completely synchronized (one-cluster) state, while a spatially organized partially synchronized state develops in networks with symmetrical coupling topology. In a previous study,17 we provided an experimental evidence for the existence of the Kuramoto-chimera state in a non-locally coupled regular (NLR) electrochemical network of anodic nickel dissolution in sulfuric acid.

In this paper, we present a detailed study of properties of the chimera patterns in the electrochemical network system. Numerical simulations with experiment-based phase models are presented, which are used as guidance for the optimization of the experimental conditions (delay in phase interaction function, network size, connectivity) for the chimera state. The experimental system has unavoidable heterogeneity (likely due to varying surface conditions) in the form of a frequency distribution of the oscillating units; to investigate the balancing effect of coupling strength for suppression of heterogeneities due to synchronization, and enhancement of heterogeneities due to the chimera effect, we characterized the features of the chimera pattern under weak, intermediate, and strong coupling strengths. The chimera pattern occurs only transiently in the experiments; the transient and long-term behaviors of the patterns are also presented. The chimera patterns are compared to a weakly heterogeneous system with classical features, and to a more heterogeneous ‘remnant chimera’ consisting of frequency clusters. Finally, the features of the chimera patterns in the electrochemical system are compared to those of the other experimentally reported patterns in chemical and physical systems.

2. Experimental and numerical methods

2.1. Experimental setup

A standard three-electrode cell consisting of a nickel working electrode array, a Hg/HgSO₄/saturated K₂SO₄ reference electrode, a platinum counter electrode, and 3 mol L⁻¹ H₂SO₄ electrolyte solution were used in the experiments (Fig. 1a). The reactor temperature was maintained at 10 °C by a circulating bath. The electrode array was made with 1 mm spacing using 1 mm diameter electrodes; the wires were embedded in epoxy so that reactions take place at the end. The electrode array was machine polished on a series of sand papers (P180 – 4000). Each wire of the electrode array was connected to a potentiostat through an external resistance. The electrodes are polarized at a constant circuit potential \( V \) which was set to 20 mV anodic to a Hopf bifurcation to obtain stable smooth electrochemical oscillation shown in Fig. 1b. Currents across the external resistances were acquired at 200 Hz data acquisition rate using a National Instruments PCI 6255 data acquisition board. A typical dataset consists of 200 oscillations with 600 data points per cycle.

Non-locally coupled regular (NLR) electrochemical reaction network. A node of a network is an oscillatory nickel dissolution reaction occurring on the surface of an electrode of the array. A network edge (link) is created with a coupling resistance (\( R \)). A capacitor parallel to each coupling resistor is added to induce some delay in coupling current.\(^{41}\) The schematic of NLR with twenty electrodes and 140 links is shown in Fig. 1c. An electrode is connected to seven of its nearest neighbors (in both directions along the chain) distributing connections symmetrically. The network consists of 140 total connections for twenty electrodes. The coupling strength between two oscillators is defined as an inverse of coupling resistance, i.e. \( K = 1/R \).

2.2. Frequency of oscillations

We use the Hilbert transform of the time series of current

\[
H(t) = \frac{1}{R} \text{PV} \int_{-\infty}^{\infty} \frac{I(t') - \langle I \rangle}{t' - t} dt',
\]

in the defining phase\(^{42}\)

\[
\phi(t) = \arctan \left( \frac{H(t)}{I(t) - \langle I \rangle} \right)
\]

\( \text{PV} \) in eqn (1) implies that the integral should be evaluated in the sense of Cauchy principle value. \( \langle \cdot \rangle \) denotes the temporal average. The frequency \( \omega \) of an oscillator is obtained from a linear fit of \( \phi(t) \) vs. \( t \).

\[
\omega = \frac{1}{2\pi} \left\langle \frac{d\phi}{dt} \right\rangle
\]

2.3. Mean field phase

The mean field phase of element \( k \) in the NLR network is defined as

\[
\Theta(t) = \arctan \frac{1}{2L + 1} \sum_{k-L}^{k+L} \exp(i\phi_k(t))
\]

where \( i \) is the complex unit, \( L = 7 \) is the radius of the coupling (the element indices are circular). The phase of an oscillator...
relative to the mean phase (phase difference) is \( \theta(t) = \phi(t) - \Theta(t) \).

2.4. Kuramoto order parameter
The amplitude of the global mean field of coupled oscillators is characterized by the Kuramoto order parameter\(^{23}\) as

\[
Z(t) = \left| N^{-1} \sum_{j=1}^{N} e^{i\phi_j(t)} \right|
\]

(5)

where \( N \) is the number of nodes of the network.

3. Results and discussion

3.1. Numerical simulations
The chimera pattern is first explored with simulations using phase models. The goal of the simulations is twofold. First, they reveal the dynamical characteristics of the chimera patterns to be identified later in the experiments. Second, there is a limitation in the experiments on the number of network nodes, number of links, and delay in the phase interaction function that can be achieved; the simulations are used to identify experimentally attainable conditions under which the chimera state can occur.

3.1.1. Model simulations with ideal phase models. Numerical simulations are performed with a ring array of \( N \) non-locally coupled phase oscillators with phases \( \phi_j(j = 1, 2 \ldots N) \) evolving according to the model description:\(^{43}\)

\[
\frac{d\phi_j}{dt} = 2\pi \Omega_j + g \sum_{k=1}^{N} M_{k,j} \Gamma(\phi_k, \phi_j)
\]

(6)

where \( \Omega_j \) is the natural frequency of oscillators, \( M \) is the connection matrix with element 1 for a connection between oscillators \( k \) and \( j \), and 0 for the absence of a connection, and \( g \) is the coupling strength. Motivated by observations of Wolfrum and Omel’chenko,\(^{31}\) we started simulations on network with a population of forty identical oscillators (\( N = 40 \)) with the phase interaction function \( \Gamma \) given by \( a \sin(\phi_k - \phi_j) + b (1 - \cos(\phi_k - \phi_j)) \). Such an interaction function can be observed with oscillatory behavior close to a Hopf bifurcation.\(^{23}\) The units are arranged in a ring; each oscillator is connected to 14 nearest neighbor oscillators (coupling range \( L = 14 \)) on each side giving a total number of 560 links.

We present simulation results of (\( N = 40, L = 14 \)) with setting \( a = 0.01101 \) and \( b = -10 \times a \). The solution of eqn (6) for identical oscillators with \( \Omega_j = 0.4 \) Hz and \( g = 0.26 \) exhibits features of the typical chimera behavior. Phase dynamics of 211 cycles after the initial transient behavior were analyzed for features of the typical chimera behavior. Phase dynamics of the population of forty identical oscillators (coupling range \( L = 14 \)) on each side giving a total number of 560 links.

3.1.2. Simulations with experiment-based phase models. The direct realization of the chimera state through the Kuramoto simulations thus reveal that with a phase model that contains small positive, phase attracting sinusoidal and large cosine harmonics in the interaction function, the chimera state can be realized. As quantitative means for the pattern characterization, we can analyze the variation in frequency and a position dependent synchronization to the mean field along the chain. The chimera state is exhibited as a transient, spatially organized partially synchronized state with a group of core elements that are synchronized, and a group of shell elements that are desynchronized.

3.1.2. Simulations with experiment-based phase models. The direct realization of the chimera state through the Kuramoto
mechanism presented in Section 3.1.1 has many experimental obstacles including

(i) The oscillators are heterogeneous, i.e., there is a distribution of the natural frequencies. Even relatively small extent of heterogeneity is capable of destroying the pattern.  
(ii) The phase interaction function in eqn (6), which was experimentally measured previously, has weak higher harmonics. The impact of these small higher harmonics on the formation of the chimera state needs to be explored.
(iii) The experimental conditions must be realized in such a manner that there is a large ratio of cosine and sine functions. The ratio in a given system depends on the properties of the oscillator (e.g., timing of inherent kinetic feedback loops, values of rate constants, and other parameters such as resistance and potential for an electrochemical system) and the type of coupling (difference or differential coupling, or through coupling delay).
(iv) The relatively large number of network links (>500) is not possible to realize in our experiments. The number of links and the number of electrodes should be kept below 200 and 80, respectively.

In the first step, our objective was to reduce the total number of links to a value that we can realize in the design of the experiments while the solution to eqn (6) exhibits features of the chimera state. A series of simulations was carried out for different network sizes and number of connections \( (N = 30, L = 11), (N = 30, L = 14), (N = 20, L = 5), (N = 20, L = 7), (N = 18, L = 7), \) and \( (N = 16, L = 5) \) with homogeneous oscillators (with \( \Omega = 0.4 \) Hz). These simulations indicated that the chimera state can robustly exist with a network size of 20 oscillators with 7 nearest neighbor coupling \( (N = 20, L = 7) \). The network is constructed from 140 total connections, which is below the experimental limit. The simulations show a chimera state similar to that observed in Section 3.1.1, but with a shorter lifetime of about 200 oscillations.

In the next step, we considered a heterogeneous oscillator system where the natural frequencies of oscillators \( (\Omega_j) \) were chosen from a set of experimental frequencies of current oscillations in Ni electrodissolution reaction (with a mean frequency of 0.3969 Hz, standard deviation of 0.1 mHz, and range of 0.5 mHz). As it was described previously, simple difference coupling between the electrodes (implemented through cross resistors, \( R \)) results in a phase model with a pure sinusoidal component. However, a differential coupling (obtained with cross-capacitors, \( C \)) gives a phase interaction function with predominant cosine components. After several experimental trials for \( RC \) combinations, we measured a phase interaction function

\[
\Gamma(\phi_k, \phi_j) = 0.00026 \pi \sin(\phi_k - \phi_j) - 0.0027 \pi [1 - \cos(\phi_k - \phi_j)]
\]

under the coupling configuration \( R = 50 \) k\( \Omega \) and \( C = 48 \) m\( \Omega \) in a system of two coupled oscillators with a 15 mHz natural frequency difference. (The experimental determination of the interaction function in the Ni/H2SO4 electrochemical system is described elsewhere.  

This interaction function has a \( |\cos(\Delta \phi)|/|\sin(\Delta \phi)\) coefficient ratio of 10; very close to the \( |a/b\) ratio in the ideal phase model with \( N = 40, L = 14 \) in Section 3.1.1.

Simulation results of the \( (N = 20, L = 7) \) NLR network are shown in Fig. 3 for heterogeneous oscillators having a natural frequency distribution range of 0.5 mHz (Fig. 3a, open circle) and coupling strength of \( g = 0.503 \). Simulated dynamics were analyzed for chimera behavior in the time frame \( t = 300–820 \) (204 cycles) when the Kuramoto order parameter was around \( Z(t) = 0.78 \) (Fig. 3b). Eight oscillators, 1–4 and 17–20, form the synchronized core with a frequency of 0.395 Hz and the rest of the oscillators, 5–16, are in the desynchronized shell. The average frequency deviation of the shell is about 5 mHz from the core oscillators; 10 times the range of natural frequency distribution. Note that even though shell oscillators such as 7–10 appear to have similar frequencies in Fig. 3a, they are not synchronized to their mean field (Fig. 3c). The same explanation can be drawn for oscillators 5–6 and 11–13 in Fig. 3a. Core-oscillators are phase locked and shell-oscillators exhibit phase slipping behavior with respect to the mean field phase during the chimera state. Oscillators become fully phase synchronized when the order parameter reaches \( Z = 1 \) as shown in Fig. 3b after about 300 oscillations.

Simulations were also carried out with the same 20-oscillator NLR network for weak coupling strength at \( g = 0.005 \) (not shown). The chimera behavior was absent.

The simulations thus reveal that the chimera state is attainable with a 20-oscillator ring network with each element coupled to 7 nearest neighbors to the left and right along the chain. The experiment-based phase models show that the chimera state is robust against heterogeneities of about 0.5 mHz. However, a sufficiently large coupling strength needs to be applied to suppress the heterogeneities. The expected lifetime of the experimental chimera state is in the order of 100 oscillations.
3.2. Experiments

All experiments were performed with a non-local regular network of 20 reaction units (electrodes or nodes) with 140 connections. The difference between the experiments was the applied coupling strength and the extent of the heterogeneities (experimentally observed as the range of natural frequency distribution) of the electrodes.

3.2.1. Chimera state of non-locally coupled smooth oscillators.

Without any added coupling, the current oscillation of each electrode has a slightly different frequency resulting in a distribution in a range of about 16 mHz (see Fig. 4a). The frequency distribution is likely caused by surface heterogeneity: the natural frequencies of each wire are reproducible between subsequent repolishing within about 1–2 mHz error; this error is smaller than the observed range, which indicates the importance of metal impurities in the course of the dissolution process. In addition, the oscillation frequency also changes slowly with time (about a few mHz per hour) and thus additional properties (spatial oxide film heterogeneities and oxide layer thickness) could also contribute to the frequency distribution. Surface heterogeneities also played a role in the dynamics of \( \text{H}_2\text{O}_2 \) reduction on the \( \text{p-CuInSe}_2 \) electrode; the inhomogeneously growing passivating surface layer formed islands, and the islands grew with time resulting in a time-transient dynamical response. Heterogeneities thus could occur for many electrochemical oscillatory systems and their extent and time variation could significantly impact the observed dynamics.

Experiments were performed by small adjustments to individual resistors \( R_{\text{ind}} \) which narrowed the natural frequency range of oscillators from 16 mHz to 0.5 mHz (see Fig. 4a, b and 5b), before tuning on coupling. This ensured that the chimera mechanism was responsible for the intensified incoherence among ‘shell’ oscillators. As the numerical simulations indicated, we employ a combination of capacitor and resistor in parallel as a coupling element so that the coupling current across the resistor is delayed by the capacitance.

When coupling was turned on, a group of synchronized ‘chimera-core’ electrodes, elements 1–4 and 17–20 in Fig. 5a, and a group of desynchronized ‘shell’ electrodes, elements 5–16 in Fig. 5a, were observed for 84 cycles. (This chimera state is the same as published previously; here we review the results for subsequent comparisons at various coupling strengths and heterogeneities.) The eight core electrodes have very similar currents (see Fig. 5a) and deviations arise outside the core region. Eight core oscillators are synchronized to a frequency of 0.389 Hz and all shell elements have lower frequencies than the core frequency. The frequency distribution in Fig. 5b reveals that shell elements are desynchronized and they form a lower semi-circle as a function of position confirming the numerical simulation results. The strong incoherence between the core and the shell domains broadens the initial 0.5 mHz frequency range to about 18 mHz. This frequency enlargement indicates the presence of the chimera symmetry breaking mechanism.

A plot of phase of each oscillator relative to the mean field phase \( \Theta(t) \) in Fig. 5d further confirms the chimera behavior of the system. Phases of core oscillators are locked to the mean field phase (therefore, phase difference is zero) while phases of the shell oscillators exhibit phase slipping behavior (time sequences of phase locking is interrupted with a relatively quick \( 2\pi \) phase slip). Note that although many shell elements have similar frequencies,
they are not synchronized to the mean field. Shell oscillators near the core display only 1–2 phase slips in 84 cycles as shown in Fig. 5c while shell elements away from the core exhibit about 4 phase slips. Fig. 5c shows the Kuramoto order parameter as a function of time; the system exhibits partial synchrony with a mean value of $Z = 0.73$. After the break-up of the chimera state the system typically approaches full synchrony with $Z = 1$ (not shown). Therefore, the chimera state in the experiment is a long transient state. The transient nature of the experimental system could arise due to drifting natural frequencies of oscillators as well; however, as it was shown numerically with the phase eqn (6) in Section 3.1, the chimera state in the given coupling configuration was an inherent transient behavior for a finite number of oscillators.

The chimera state was stable for about 80–100 cycles and was observed in 9 out of 14 experiments. Most successful attempts (6/9) were made during the first four hours when natural frequency-drifts of oscillators were minimal. In the remaining five experiments, most (4/5) were done after four hours, less than 5 oscillators exhibited synchronization in 100 cycles.

The experiments thus confirm that the chemical reaction system, within experimental limitations, exhibits a very robust partially synchronized state with behavioral characteristics identical to the predicted chimera state. The coupling induces co-existing synchronized and desynchronized states even in a symmetrical network. Although the lifetime of the chimera state is relatively short (about 100 oscillations), it is comparable to that obtained from numerical simulations. The most unexpected feature of the partially synchronized state is that the coupling induces synchronization among core elements; nonetheless, the very same coupling enhances frequency differences for shell elements. Because the coupling among the oscillators was induced with current through a parallel resistance/capacitance element, the coupling signal has a timescale of $RC = 2.35$ s. This coupling time-scale matches the time scale (period) of the oscillatory process (2.5 s). Similar time-scale matching has been observed in the chimera state observed with the coupled pendulum system. The electrochemical experiments thus confirm the idea proposed in the pendulum chimera system about the prevalence of the mechanical analogy of development of a chimera state as a competition between different synchronization modes in a coupling parameter region centered around the resonance curve where the uniform synchronization breaks up.

3.2.2. Chimera state at weak coupling strength. A weak form of chimera behavior was observed with a 17% ($R = 600 \, \text{k}\Omega$, $C = 4.7 \, \mu\text{F}$) decrease in the coupling strength in comparison to the chimera state presented in the previous section. To minimize practical difficulties the capacitance $C$ was kept constant in the experiments. The chimera behavior is obtained for 100 cycles having six electrochemical oscillators (1, 2, 17–20) in the synchronized core with a frequency of 0.403 Hz; the rest of the oscillators are in the desynchronized shell with a frequency distribution range of 14 mHz as illustrated in Fig. 6a. Weakening the coupling strength thus diminishes the size of the core of the chimera state. A well-defined semi-circle frequency distribution is not observed for weak coupling. This chimera state was obtained when the Kuramoto order parameter maintained an average value of $Z = 0.85$ as shown in Fig. 6b. In the experiment with stronger coupling in Section 3.2.1, we observed that the number of phase slips increases gradually from both edges of the core to the middle of the shell. However, under weak coupling, we can see that from the edge electrode 17 (no phase slip) of the chimera core, there are 1–2 phase slips for chimera shell elements 16–12, three phase slips for elements 11–10, and four phase slips for element 8; the trend is disrupted by element 9 which exhibits only one phase slip (see Fig. 6c).

Starting from the other core edge, electrodes 3–7 all exhibit 2–3 phase slips. We thus see that the number of phase slips from the chimera core to the center of the shell elements increases in a statistical manner and exhibits strong asymmetry; these results indicate that the pattern is affected by the heterogeneity of the oscillators that are not fully suppressed at the weak coupling. The spatially ordered partially synchronized state was difficult to obtain with weak coupling; the presented example is the only experiment that produced the chimera state in seven trials. In other experiments, electrochemical oscillators were weakly synchronized without apparent spatial organization during a period of 100 cycles.

3.2.3. Chimera state at strong coupling strength. The chimera state was observed for 190 cycles with a relatively strong coupling resistance $R = 390 \, \text{k}\Omega$ (capacitance $C = 4.7 \, \mu\text{F}$). Fig. 7a shows frequencies of oscillations. The chimera state at this strong coupling consists of a relatively large domain (1–7, 17–20) of the core elements. The desynchronized shell elements deviate only about 10 mHz from the synchronized core frequency of 0.381 Hz. Although the characteristic semi-circle frequency variation in the frequencies of the shell elements is not noticeable, the system displays the prominent feature of the chimera state: the coexistence of synchronized and desynchronized groups with

![Fig. 6](image_url)
a clear separation of shell frequencies from the core frequencies. The center of the shell of the chimera state has the lowest frequency and the most (5) phase slips; the element at the left edge of the shell state exhibits only one phase slip. The other shell elements exhibit 2–3 phase slips (see Fig. 6c). The network exhibits chimera behavior when the Kuramoto order parameter fluctuates between 1 and 0.6 (see Fig. 7c).

With very strong coupling (e.g., $R = 40 \, \text{k}\Omega$, $C = 33 \, \mu\text{F}$), all 20 oscillators formed one group attaining full phase synchronization.

The experiments thus show that the chimera state can be observed for a range of coupling strength. The most robust chimera pattern is observed at an intermediate coupling strength where there is a balance between three major factors that include heterogeneity suppression, core and shell element differentiation, and full synchronization. Upon increasing the coupling strength the domain of the core of the chimera patterns increases but remains within about 30–55% of the total population.

### 3.2.4. Long-term behavior of the NLR electrochemical network

The experimental chimera state, which closely resembles the numerical simulation, lasted under intermediate coupling strength for about 100 oscillatory cycles. A long-term behavior (including the initiation and the termination) of the dynamics under intermediate coupling strength conditions $R = 499 \, \text{k}\Omega$ ($C = 4.7 \, \mu\text{F}$) is shown in Fig. 8.

The system is initiated from a desynchronized state with low value of the Kuramoto order parameter at $t = 0$. When the coupling is turned on, the oscillators synchronize and quickly approach a state in which $Z$ is close to 1 at $t = 250 \, \text{s}$ (Fig. 8b). This fully synchronized state collapses to a chimera behavior at $t = 600$; thereby, the average Kuramoto order parameter decreases to 0.87.

Nine electrodes (4–7, 9–13) form the synchronized core with a frequency of 0.395 Hz as shown in Fig. 8a. (Element 8 exhibits only one phase slip during the time interval for the chimera state.) The chimera state was sustained for the next 215 cycles. (The conditions for the chimera state are the same as those in Fig. 5, however, now we analyze the chimera state for more than twice as long time interval − 215 cycles vs. 84 cycles). The maximum frequency difference between shell and core oscillators is 13 mHz. The semi-circle distribution of frequencies is not apparent compared to that of the short-term chimera behavior under the intermediate coupling strength. Inherent drifts in natural frequencies adversely affect the formation of the chimera state since increasing the heterogeneity in frequencies prevents the formation of the chimera state. Phases of oscillators relative to the mean field phase are shown for the chimera state in Fig. 8c.

In the experiments the chimera state collapses after some time; this collapse often occurs through the formation of frequency clusters in which the frequency of the twenty electrodes is split into several small domains (phase synchronization within a domain) as shown in Fig. 9. In this state, two types of frequency clusters can be observed. A chimera-core-like cluster is formed with oscillators 1–3, 6, and 17 (Fig. 9a-black circles); these elements are entrained to both the mean field and to each other. For example, electrode 1 is in-phase with both the mean field (Fig. 9c) and another chimera core element 2 (see Fig. 9b). (Note that because of a phase slip that occurred before the formation of the frequency clusters, the phase difference between elements 1 and 2 is $2\pi$ rad instead of zero.) In the other type of frequency cluster groups, the elements are not entrained to the mean field, but they are entrained to each other. Such clustering occurs among oscillators 12–15 with a common frequency of 0.371 Hz. For instance, the phase difference between oscillators 13 and 14, shown in Fig. 9b, wiggles between 0 and $2\pi$ rad, however, the phase difference is bounded and does not grow; this leads to phase synchronization between elements 13 and 14. However, phase evolutions of both 13 and 14 are not
This journal is (partially) suppress the relatively large heterogeneities (15 mHz strictly defined by the Kuramoto conditions. in heterogeneous oscillator network. typically a frequency clustered state. during a long-term experiment, and the final state is state (100 cycles (1 the chimera state is typically observed approximately for frequency clusters. In some long-term experiments with intermediate coupling, the chimera state is typically observed approximately for 100 cycles (1 > Z > 0.7) and it collapsed to a fully synchronized state (Z = 1) for about another 100 cycles. This alternation between chimera and fully synchronized states appears in 2–3 occasions during a long-term experiment, and the final state is typically a frequency clustered state. 3.2.5. Remnant chimera state with frequency clustering in heterogeneous oscillator network. Heterogeneous natural frequencies do not facilitate the formation of the chimera state strictly defined by the Kuramoto conditions. The typical behavior of a heterogeneous population (with multimodal frequency distribution) with global coupling is the formation of frequency clusters. However, under conditions favorable for chimera states, we found that the formation of the frequency clusters is affected by chimera dynamics through a state quite similar to the terminal state shown in Fig. 9. Strong coupling strength at R = 390 kΩ (C = 4.7 μF) was needed to (partially) suppress the relatively large heterogeneities (15 mHz range in natural frequency in Fig. 10) of the oscillating elements. Under these conditions, five frequency clusters were obtained as illustrated in Fig. 10a and b; the clusters have frequencies 0.3966 Hz ± 0.1 mHz (elements 1, 2, 14–16), 0.3940 Hz ± 0.1 mHz (elements 4, 7, 9, 17–20), 0.3913 Hz ± 0.3 mHz (elements 3, 8), 0.3887 Hz ± 0.4 mHz (elements 11–13), and 0.3860 Hz ± 0.4 mHz (elements 5 and 10). The state can be considered as a ‘remnant’ chimera state where the core of the chimera consists of elements 1–2, 4, 7, 9, and 14–20. Within the core, elements 1, 2, and 14–16 are synchronized to each other and to their mean field while the remaining elements exhibit only about two phase slips in a relatively long time of 200 oscillations (see Fig. 10d). Note that a large domain of the chimera core consists of contiguous elements 1–2 and 14–20; the other elements (4, 7, and 9) are in spatially isolated regions away from the major domain. (We did not find evidence that the core or the shell of the chimera state was pinned to heterogeneities in an obvious manner; for example, half of the chimera core elements had similar natural frequencies, while the other half had largely different natural frequencies.) The noisy chimera shell (elements 3, 5–6, 8, and 10–13) is formed from 4 frequency clusters. In each cluster, the elements are synchronized to each other but not to the mean field; the typical size of the cluster varies between 1 and 3 elements. The elements in these frequency clusters can consist of either contiguous elements (e.g., elements 11–13) or from spatially distinct units (e.g., elements 3 and 8). The Kuramoto order parameter fluctuates strongly between 0.5 and 1 as shown in Fig. 9 and Fig. 10.
Fig. 10c; such fluctuating order parameters are often observed with frequency clustering.\(^{56}\)

4. Conclusions

In summary, we obtained spatially organized partial synchronization in a non-locally coupled electrochemical network through the Kuramoto chimera mechanism.\(^{22,24,26}\) An experiment-based phase modeling technique\(^{34}\) was used to optimize the network topology (20 units on a ring, coupled to 7 nearest neighbors on both sides) and the time scale of the coupling signal (2.5 s), which matched the period of oscillations (about 2.35 s). The chimera state was most robustly observed at intermediate coupling strength where the inherent heterogeneities were suppressed by the coupling while the chimera symmetry breaking mechanism was responsible for desynchronization of the shell elements. Within the coupling range that supported formation of the chimera state, the core (synchronized) portion of the pattern increased in size with an increase in the coupling strength. Increasing the heterogeneity among electrochemical oscillators did not favor developing the chimera state. However, an important fingerprint of chimera behavior was found with the heterogeneous elements: in one of the frequency clusters, which are typically composed of contiguous elements, the units are not only synchronized among each other, but also to their corresponding mean fields. This finding could facilitate the identification of ‘remnant’ or ‘noisy’ chimera states (e.g., in biological system) where the inherent heterogeneities are relatively strong. We note, however, the calculation of a quantity for the extent of synchronization to the mean field requires the complete identification of the network topology, which can be a tedious task in systems where the links are difficult to visualize, e.g., with cellular systems.\(^{57}\)

The chimera pattern obtained with the electrochemical experiment can be compared to those obtained with the BZ bead system\(^{15,48}\) and the silicon dissolution system.\(^{38,39}\) The chimera state with the BZ bead system was observed with a spatially distributed illumination feedback that implemented a NLR or a two-group network of elements.\(^{54,48}\) A major conclusion of the BZ bead experiments is that the chimera state has features that differ significantly from those obtained with phase-oscillator models. Most notably, the presence of clustering (which was reported with global feedback) could play an important role in the desynchronization mechanism of the shell elements. The chimera-like state with the silicon dissolution experiments\(^{38,39}\) arises as a competition between various cluster states and sizes, and thus can be observed with pure global coupling. In addition, the system should have sufficiently large nonlinearity (most likely through the coupling mechanism) to support the co-existence of domains of the vastly different types of cluster states.\(^{38}\) In our experiments the mechanism for the chimera state could be attributed to the original Kuramoto mechanism;\(^{34}\) the mechanism can be clearly identified because of the existence of experiment-based, predictive phase models for electrochemical oscillators.\(^{24,49}\) The experiments presented with the Ni electro-dissolution system here are examples of pattern formation uniquely induced by the presence of a network, and not a representation of a dynamical behavior (e.g., clustering) in the presence of a network. Categorization of the different types of chimera-like behaviors is an important future direction for physical, chemical, and biological oscillatory systems.

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