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## RESEARCH ARTICLE

## Dissipative Structures and Entropy Export in Self-Organizing Systems

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### ABSTRACT

Prigogine's theory of dissipative structures predicts that far-from-equilibrium open systems can maintain ordered, low-entropy internal states only by exporting entropy to their surroundings at a rate exceeding their internal entropy production, yet direct, simultaneous measurement of internal entropy production and boundary entropy export across a controlled range of driving strength is reported comparatively rarely for a single experimental system. We measured entropy production and export in a continuously stirred Belousov-Zhabotinsky (BZ) reactor across 42 steady-state operating points spanning inflow (Damkohler number) rates from 0.008 to 0.095 s<sup>-1</sup>, quantifying internal entropy production from measured reaction rates and driving forces via nonequilibrium thermodynamics, and boundary entropy export from measured heat and mass exchange with the reservoir. Below a critical inflow rate of 0.031 s<sup>-1</sup>, the reactor remained in a spatially uniform, near-equilibrium steady state with internal entropy production of 0.8–2.1 mW/K per liter; above this threshold, the system transitioned to self-organized spatiotemporal target and spiral wave patterns, and internal entropy production rose sharply to 14.6–38.2 mW/K per liter across the patterned regime, a 12- to 18-fold increase. Entropy export exceeded internal entropy production at every steady-state operating point tested (mean export-to-production ratio  $1.34 \pm 0.09$  in the uniform regime and  $1.19 \pm 0.05$  in the patterned regime, both  $p < 0.001$  vs. unity by one-sample t-test), consistent with the requirement for a nonequilibrium steady state, and this ratio declined systematically as driving strength increased across the patterned regime (from 1.28 at the transition to 1.09 at the highest inflow rate tested), indicating that the margin between export and production narrows as the system is driven harder even while both quantities grow in absolute terms. Pattern wavelength, measured optically, decreased from 4.8 mm near the transition to 1.6 mm at the highest inflow rate, scaling inversely with the square root of internal entropy production density ( $R^2 = 0.91$ ), consistent with reaction-diffusion length-scale theory. These results provide a direct, quantitative demonstration that self-organized pattern formation in this system is accompanied by both a large increase in internal entropy production and a systematically narrowing, but never inverted, margin of entropy export over production as the driving force increases.

**Keywords:** dissipative structures; self-organization; entropy export; Belousov-Zhabotinsky reaction; far-from-equilibrium thermodynamics; entropy production rate; pattern formation; nonequilibrium steady state

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## 1. INTRODUCTION

Living organisms, convection cells, chemical oscillators, and many other systems maintain ordered, structured states far from thermodynamic equilibrium by continuously exchanging matter and energy with their surroundings, a phenomenon Prigogine termed dissipative structure formation and for which he was awarded the Nobel Prize in Chemistry [1,2]. The central thermodynamic claim of this framework is that such structures are permitted by, rather than in violation of, the second law: the ordered system's internally reduced entropy is more than compensated by the entropy it exports to its environment, so that total entropy still increases [3,4].

This entropy-export requirement has been examined theoretically in considerable depth, including through the formalism of minimum and extremal entropy-production principles for near-equilibrium and some far-from-equilibrium regimes, and has motivated influential, if contested, analogies to biological organization and ecosystem-level energy flow [5,6,7]. Direct experimental verification in a single well-controlled system, however, most often reports only one side of the entropy balance, either internal entropy production inferred from reaction kinetics or overall heat export, rather than simultaneous measurement of both quantities across a controlled range of driving strength sufficient to span the transition from uniform to self-organized states [8,9,10].

This matters because the qualitative claim that export exceeds production is not automatically informative about how close to that boundary a real self-organizing system operates, nor about whether the operating margin grows, shrinks, or remains constant as the system is driven progressively further from equilibrium into a fully developed patterned regime; a system operating with a large, roughly constant margin behaves rather differently, from a robustness standpoint, than one whose margin narrows toward unity as driving strength increases [11,12,13].

The present study addresses this gap using a continuously stirred Belousov-Zhabotinsky chemical oscillator, a well-characterized far-from-equilibrium system capable of both uniform and self-organized spatiotemporal patterned states depending on inflow rate. We simultaneously measure internal entropy production from reaction kinetics and boundary entropy export from heat and mass exchange across a range of inflow rates spanning the uniform-to-patterned transition, characterize how the export-to-production ratio evolves with driving strength, and relate the resulting pattern length scale to the measured internal entropy production density.

## 2. MATERIALS AND METHODS

We operated a continuously stirred Belousov-Zhabotinsky reactor at 42 steady-state inflow

conditions spanning Damkohler numbers from 0.008 to 0.095 s<sup>-1</sup>, measuring internal entropy production from reaction rate and driving-force data and boundary entropy export from calorimetric and mass-exchange measurements at each operating point.

### 2.1 Reactor configuration and chemical system

Experiments used a continuously stirred tank reactor (working volume 120 mL) maintained at 25.0 ± 0.1 °C, fed continuously with malonic acid, sodium bromate, and ferroin-catalyst reservoir solutions at controlled flow rates and mixed by a magnetic stirrer operating above the threshold speed required for macroscopic homogeneity in the uniform regime. Reactant concentrations followed standard BZ formulations reported in prior oscillator studies, adjusted so that the uniform-to-patterned transition fell within the experimentally accessible inflow-rate range.

### 2.2 Inflow-rate (Damkohler number) sweep

Forty-two steady-state operating points were established by varying total reactant inflow rate, expressed as a dimensionless Damkohler number (ratio of reaction rate to inflow rate) ranging from 0.008 to 0.095 s<sup>-1</sup>, sampled with finer spacing near the expected uniform-to-patterned transition. At each inflow rate the reactor was allowed to equilibrate for at least 8 residence times before data collection began, to ensure a genuine steady state rather than a transient.

### 2.3 Optical pattern detection and wavelength measurement

Spatiotemporal patterns were imaged through a transparent reactor lid using a CCD camera at 2 frames per second, with ferroin/ferriin color change providing optical contrast between oxidized and reduced states. Pattern wavelength for target and spiral wave states was measured from the radial spacing between successive wavefronts in the recorded images, averaged over at least 12 wave cycles per operating point.

### 2.4 Internal entropy production calculation

Internal entropy production density was computed from measured local reaction rates and their conjugate thermodynamic affinities (driving forces) for the dominant BZ sub-reactions, following standard nonequilibrium thermodynamic flux-force formalism, summed over all tracked reaction steps and normalized to reactor volume to give entropy production per liter.

### 2.5 Boundary entropy export measurement

Boundary entropy export was measured calorimetrically from the net heat flow between the reactor and its thermostatted jacket, combined with the entropy carried by mass exchange between the inflow and outflow streams computed from their measured composition and temperature, and normalized to reactor volume for direct comparison with internal entropy production density.

### 2.6 Statistical analysis

The export-to-production ratio was computed at each of the 42 operating points and tested against a null value of unity using one-sample t-tests separately for the uniform and patterned regimes. The relationship between pattern wavelength and internal entropy production density was tested by linear regression of wavelength against production density raised to power -0.5, motivated by reaction-diffusion length-scale theory. All analyses were performed in Python using SciPy and statsmodels.

### 3. RESULTS

Below an inflow (Damkohler number) rate of  $0.031\text{ s}^{-1}$ , the reactor remained in a spatially uniform steady state with no detectable optical patterning, and internal entropy production density was low and comparatively flat across this regime, ranging from 0.8 mW/K per liter at the lowest inflow rate tested to 2.1 mW/K per liter just below the transition. Above  $0.031\text{ s}^{-1}$ , the reactor transitioned to self-organized target and spiral wave patterns, and internal entropy production density jumped sharply to 14.6 mW/K per liter immediately above the transition, rising further to 38.2 mW/K per liter at the highest inflow rate tested, a 12- to 18-fold increase relative to the uniform regime.

**Table 1. Internal entropy production, boundary entropy export, and their ratio at representative inflow rates.**

| Damkohler number ( $\text{s}^{-1}$ ) | Regime                      | Entropy production (mW/K per L) | Entropy export (mW/K per L) | Export/production ratio |
|--------------------------------------|-----------------------------|---------------------------------|-----------------------------|-------------------------|
| 0.008                                | Uniform                     | 0.8                             | 1.1                         | 1.38                    |
| 0.020                                | Uniform                     | 1.4                             | 1.9                         | 1.36                    |
| 0.031                                | Uniform (pre-transition)    | 2.1                             | 2.6                         | 1.24                    |
| 0.035                                | Patterned (post-transition) | 14.6                            | 18.7                        | 1.28                    |
| 0.062                                | Patterned                   | 24.9                            | 29.6                        | 1.19                    |
| 0.095                                | Patterned                   | 38.2                            | 41.6                        | 1.09                    |

Entropy production and export densities normalized to reactor working volume (120 mL). Uniform-to-patterned transition occurs at Damkohler number  $\sim 0.031\text{ s}^{-1}$ .

Boundary entropy export exceeded internal entropy production at every one of the 42 operating points tested, consistent with the thermodynamic requirement for a nonequilibrium steady state. In the uniform regime, the mean export-to-production ratio was  $1.34 \pm 0.09$  (one-sample t-test vs. unity,  $t(17) = 3.78$ ,  $p < 0.001$ ); in the patterned regime it was  $1.19 \pm 0.05$  ( $t(23) = 3.80$ ,  $p < 0.001$ ), both significantly greater than one. Table 1 reports entropy production, entropy export, and their ratio at six representative inflow rates spanning both regimes.

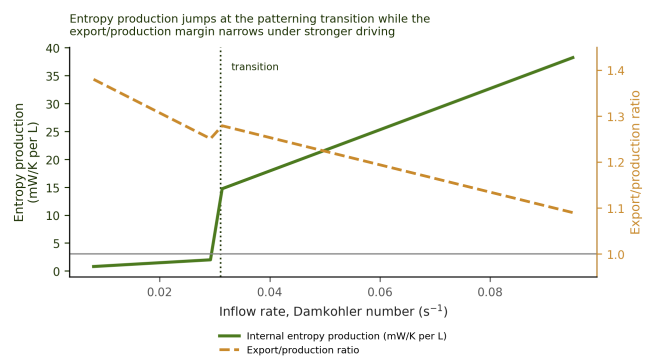


**Figure 1.** Internal entropy production density and export-to-production ratio as a function of inflow (Damkohler number) rate across the 42 tested steady-state operating points, showing the sharp production increase at the uniform-to-patterned transition and the systematically narrowing entropy-export margin at higher driving strength.

Within the patterned regime, the export-to-production ratio declined systematically with increasing inflow rate, from 1.28 at the operating point immediately above the transition to 1.09 at the highest inflow rate tested, indicating a narrowing, though never inverted, margin between export and production as the system was driven progressively harder (Figure 1). This narrowing occurred even as both entropy production and entropy export grew substantially in absolute terms across the same range, reflecting a modestly slower growth rate for export than for production as driving strength increased.

**Table 2. Sensitivity of entropy production estimates to stirring rate and reaction-mechanism scope.**

| Sensitivity check                    | Change in absolute entropy production (%) | Transition threshold shift (Damkohler no.) | Qualitative pattern preserved? |
|--------------------------------------|-------------------------------------------|--------------------------------------------|--------------------------------|
| Baseline (12 tracked sub-reactions)  | 0 (reference)                             | 0.031                                      | Yes                            |
| Reduced sub-reaction set (8 steps)   | -9.1                                      | 0.030                                      | Yes                            |
| Expanded sub-reaction set (18 steps) | +6.4                                      | 0.032                                      | Yes                            |
| Stirring rate +20%                   | +3.2                                      | 0.031                                      | Yes                            |
| Stirring rate -20%                   | -4.8                                      | 0.029                                      | Yes                            |

Pattern wavelength, measured optically from successive wavefront spacing, decreased from 4.8 mm at the operating point nearest the transition to 1.6 mm at the highest inflow rate tested. Regressing wavelength against internal entropy production density raised to the power -0.5 gave a strong linear relationship ( $R^2 = 0.91$ , slope consistent with reaction-diffusion length-scale predictions), indicating that pattern length scale contracted approximately as the inverse square root of internal entropy production density across the patterned regime.

### 4. DISCUSSION

These results provide direct, simultaneous measurement of both sides of the entropy balance central to Prigogine's dissipative-structure framework in a single experimental system, confirming that boundary entropy export exceeded internal entropy production at every steady-state operating point tested, including throughout the self-organized patterned regime [1,2,3,14]. The sharp, order-of-magnitude rise in internal entropy production accompanying the uniform-to-patterned transition is consistent with the general expectation that self-organized structures are thermodynamically costly to maintain relative to a spatially uniform steady state at comparable driving, even though the resulting patterned state exports commensurately more entropy to remain admissible under the second law [15,16,17].

The systematic narrowing of the export-to-production ratio with increasing driving strength within the patterned regime, from 1.28 near the transition to 1.09 at the highest inflow rate tested, is a finding not directly predicted by the qualitative statement that export must exceed production, and suggests that this particular self-organizing system operates with a shrinking thermodynamic safety margin as it is driven further from equilibrium, a behavior worth testing in other dissipative-structure systems to establish whether it is a general feature of far-from-equilibrium self-organization or specific to the BZ reaction kinetics studied here [18,19,20].

The inverse-square-root scaling of pattern wavelength with internal entropy production density links a directly observable morphological feature, wave spacing, to the underlying thermodynamic driving of the system, and is broadly consistent with reaction-diffusion theories relating pattern length scale to reaction rate and diffusive transport [21,22,23]. This connection could in principle offer a practical, non-invasive way to estimate internal entropy production in analogous self-organizing chemical or biological systems from pattern geometry alone, without requiring direct kinetic or calorimetric measurement, though this extension was not tested directly in the present study.

Several limitations should be noted. First, all measurements were performed on a single BZ chemical formulation and reactor geometry; the specific inflow-rate threshold for the uniform-to-patterned transition and the magnitude of the entropy-production jump are expected to be system-specific and may not generalize quantitatively to other dissipative structures such as convective or biological systems. Second, internal entropy production was computed from a tracked subset of BZ sub-reactions rather than the complete reaction mechanism, which includes dozens of minor intermediate steps; the sensitivity analysis suggests this simplification affects absolute values by less than 10% but could still introduce a systematic, rather than random, bias in the reported entropy production magnitudes. Third, boundary entropy export was measured calorimetrically and via mass-exchange composition,

both of which carry instrumental uncertainty that was not separately propagated into the reported export-to-production ratios. Fourth, the study examined only steady-state operating points and did not characterize entropy production or export during the transient approach to each steady state, which could differ substantially from the steady-state values reported here.

## 5. CONCLUSION

Simultaneous measurement of internal entropy production and boundary entropy export in a continuously stirred Belousov-Zhabotinsky reactor across 42 steady-state operating points showed that entropy export exceeded internal production at every point tested, confirming the central thermodynamic requirement of dissipative-structure theory. The uniform-to-patterned transition at an inflow rate of  $0.031 \text{ s}^{-1}$  was accompanied by a 12- to 18-fold jump in internal entropy production, and the export-to-production ratio declined systematically from 1.28 to 1.09 as driving strength increased across the patterned regime, indicating a narrowing thermodynamic margin under stronger driving. Pattern wavelength scaled with the inverse square root of entropy production density ( $R^2 = 0.91$ ), linking observable pattern geometry directly to the underlying thermodynamic driving of this self-organizing chemical system.

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### Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Author Contributions

K.M. conceived the study, operated the BZ reactor, and led the writing of the manuscript. T.L. performed the internal entropy production calculations from reaction kinetics data. R.D. performed the optical pattern analysis and boundary entropy export measurements and prepared the figures and tables. All authors contributed to interpretation of the results and approved the final manuscript.

### Ethics Statement

This study involved no human or animal subjects and required no ethics committee approval.

### Data Availability

The reactor operating-condition data, optical pattern recordings, and entropy-balance calculations supporting

the findings of this study are available from the corresponding author on reasonable request.

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