

Recurrence of Segregation in Continuously Agitated Vessels: 512-Hour Optical Evidence, a Regime Taxonomy, and a Recovery Protocol

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Abstract

Stirring is treated as a one-way operation. A spoon turned through a cup of coffee distributes the milk, and no quantity of further stirring is expected to bring it back. We report that for laminar vessels this is false, and that the return is measurable on a timescale of days. A stirred vessel is a bounded incompressible flow, so its stirring map preserves volume; a volume-preserving map on a finite measure space recurs. Because mixedness is scored not against the continuum but against an assay of finite resolution, the “unmixed” state is a set of positive measure, and Kac’s lemma converts recurrence from a limit statement into a number of impeller revolutions. For our geometry the predicted mean first return is 5.3×10^5 revolutions, or 146 h. Four studies follow. In 216 vessels agitated continuously for up to 512 h, the intensity of segregation falls to a median 0.021 within 6.2 min, plateaus, and then rises: median first recurrence 148 h (IQR 87 to 233, observed range 22 to 506; eleven vessels right-censored). A survey of 12,412 batch agitation records from 41 plants finds a median agitation duration of 22 min and no batch exceeding 29 h, so that the fraction of industrial batches ever reaching recurrence is 6×10^{-5} : the effect lies outside the range of durations run in production rather than having been overlooked inside it. In a randomised trial of 90 waste batches, agitation continued into the recurrence window recovered the light component at 94.1% purity (95% CI 91.8–96.4) using 0.9 kW h/t against 74 kW h/t for thin-film distillation. Recurrence is confined to $Re \lesssim 60$, where an island volume fraction $\phi_{isl} = 0.62$ shields striations from diffusive erasure; above $Re \approx 400$ no return occurs and the order is destroyed. We conclude that homogeneity in a laminar vessel is a transient state, that a century of turbulent-mixing practice has destroyed recoverable order in the first minutes of every batch, and that separation of a miscible pair is available for impeller shaft work alone.

Plain-language note. Mixing is assumed to run in one direction only: milk stirred into coffee stays there. We find that in thick liquids stirred slowly the two components come apart again if agitation simply continues for long enough — about six days in our vessels. Production plants do not see this because no plant agitates a batch for six days. The effect can be used to separate two miscible liquids without heat.

Keywords: chaotic advection; intensity of segregation; Poincaré recurrence; laminar mixing; KAM islands; solvent recovery

1 Introduction

Vessel 12 in No. 3 Blending House at Coldharbour Works holds 6.2 m³ and turns a four-blade paddle at 58 rpm. On the afternoon of 24 December it was charged with an alkyd let-down and a pigment paste, brought to temperature, and started; the shift that would have discharged it was not rostered, the works having closed that evening for nine days. The agitator was never stopped. When the blending house reopened on 2 January the sight glass showed three bands.

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The batch was scrapped. The deviation report, closed eleven days later, recorded the probable cause as failure of the impeller shaft coupling, which is the ordinary explanation for a stratified vessel and is almost always right. In this case it was not. The coupling was intact on inspection. The motor-hours logger recorded continuous rotation at 58.1 rpm for the whole of the closure, and the power-draw trace was flat to within 2%, with none of the step change that a decoupled shaft produces. The vessel had been stirred, without interruption, for 216 h, and it had come apart.

The literature offers no room for this. Mixing is described everywhere as a monotone approach to homogeneity: the intensity of segregation I falls from unity toward zero, and once it has fallen it stays down [3, 1]. That description is well supported over the durations for which it has been tested. Those durations are short. The longest continuous laminar agitation we can find in any industrial record — in the published duty-cycle surveys [6] and in the 12,412-batch archive of §6 alike — is 29 h, and the longest in the experimental literature is 9 h [11]. Vessel 12 ran for twenty-four times the latter.

Recurrence is not a new hypothesis. It is an old theorem which the mixing literature does not apply. A stirred vessel of fixed volume, filled and incompressible, is acted on by a volume-preserving map [7]; a volume-preserving map on a space of finite measure returns almost every configuration arbitrarily close to itself [10]. That the return time might be astronomical is the usual and sufficient reason for ignoring this. The usual estimate, however, is made against the continuum, and no mixedness index is ever computed against the continuum. It is computed on a coarse grid, at a resolution fixed by the instrument. Against that grid the unmixed state is not a point of measure zero but a set of positive measure, and Kac's lemma [5] then returns a number of revolutions rather than a limit statement.

For the geometry of our test rig the number is 146 h. Vessel 12 came apart at 216 h.

The remainder of the paper is organised as follows. Section 3 sets out the recurrence argument, the Kac estimate, and the one condition under which molecular diffusion does not erase the pattern before it returns. Section 4 describes a rig built to hold 216 vessels in continuous laminar agitation for 512 h. Section 5 reports the resulting segregation histories, in which I falls, plateaus, and then rises. Section 6 accounts for the absence of the effect from a century of plant records. Section 7 reports a randomised trial in which the effect is used to separate a miscible waste pair with no heat input. Section 8 bounds the phenomenon in Reynolds number and gives the regime taxonomy. Section 9 sets out the principal limitation, which is that the recurrence time of an individual vessel cannot be predicted.

Nomenclature

C	impeller off-bottom clearance (m)	n	impeller speed (1/s)
c	residual island shear rate (1/s)	n_t	number of island families (–)
D	vessel internal diameter (m)	Pe	Péclet number (–)
D_m	molecular diffusivity (m ² /s)	Re	impeller Reynolds number (–)
d	impeller diameter (m)	s	striation thickness (m)
H	fill height (m)	T	stirring map (–)
I	intensity of segregation (–)	δ	assay phase window (rad)
ℓ	assay cell side (m)	λ	Lyapunov exponent (1/s)
N	number of assay cells (–)	μ	normalised volume measure (–)
ζ	island halo coefficient (–)	ϕ_{isl}	island volume fraction (–)

2 Related work

Three literatures bear on the result. None of them reports it.

Mixedness as a monotone quantity. The intensity of segregation [3] remains the standard measure of batch mixedness, and its standard use is to fit a decay: $I(t) = \exp(-t/\tau_m)$, with the mixing time τ_m reported as the design output. The functional form forbids return by construction. Where a fitted $I(t)$ has been observed to depart upward from the exponential, the departure is attributed to instrument drift, to settling of a density-mismatched tracer, or to evaporative skinning at the free surface [1], and the run is discarded. All three artifacts were eliminated by construction in the present work (§4). The upward departure remained.

Chaotic advection and transport barriers. That laminar stirred vessels contain invariant regions which exchange no material with the bulk is established [9]. The islands are usually treated as a nuisance

— dead volume to be designed out with baffles — and the design literature accordingly reports island fractions for *baffled* vessels, where they are small. Unbaffled paddle vessels, which remain the common blending-house arrangement, have been measured far less often. Our own section planimetry (§3) gives $\phi_{\text{isl}} = 0.62 \pm 0.04$ for the geometry used here.

Reversibility demonstrations. The corn-syrup demonstration, in which a dye filament stretched by a rod is recovered by turning the rod backwards, is old and well understood [2]. It is not the present claim, and the difference matters. That demonstration recovers the initial state by *reversing* the forcing. We report recovery under *continued* forcing, in one direction, at constant speed, with no operator intervention of any kind. The mechanism here is recurrence, not reversibility.

What has not been done. We could locate no study, industrial or academic, that maintained a laminar vessel under continuous agitation for more than 29 h [6, 11]. The absence is a property of the equipment rather than an oversight of the literature: agitation duty cycles are set by throughput, and agitation past homogeneity adds nothing to the product.

3 Theory

3.1 Setup

Let $\Omega \subset \mathbb{R}^3$ be the filled vessel volume, and let $\mathbf{u}(\mathbf{x}, t)$ be the velocity field, incompressible ($\nabla \cdot \mathbf{u} = 0$) and periodic in time with the impeller period $T_{\text{rev}} = 1/n$. Write φ_t for the flow map and

$$T = \varphi_{T_{\text{rev}}} : \Omega \rightarrow \Omega \quad (1)$$

for the *stirring map*, the material displacement produced by one impeller revolution. Let μ be the volume measure on Ω , normalised so $\mu(\Omega) = 1$.

Lemma 1 (Measure preservation). *T preserves μ : for every measurable $E \subseteq \Omega$, $\mu(T^{-1}E) = \mu(E)$.*

Proof. Immediate from $\nabla \cdot \mathbf{u} = 0$ and the transport theorem [7]: the Jacobian determinant of φ_t satisfies $\partial_t \det \nabla \varphi_t = (\nabla \cdot \mathbf{u}) \det \nabla \varphi_t = 0$, so $\det \nabla \varphi_t \equiv 1$. $\square$

A charge is described by a concentration field $\theta : \Omega \rightarrow [0, 1]$, advected as $\theta_k = \theta_0 \circ T^{-k}$. No mixedness assay resolves θ itself. Every assay integrates it over cells.

Definition 1 (Assay coarse-graining). Fix a partition $\mathcal{P} = \{c_1, \dots, c_N\}$ of Ω into N congruent cells of side ℓ , and set $\bar{\theta}_i = \mu(c_i)^{-1} \int_{c_i} \theta \, d\mu$. The intensity of segregation is

$$I(\theta) = \frac{1}{\sigma_0^2} \frac{1}{N} \sum_{i=1}^N (\bar{\theta}_i - \bar{\theta})^2, \quad \sigma_0^2 = \bar{\theta}(1 - \bar{\theta}), \quad (2)$$

so that $I = 1$ for a fully segregated charge and $I = 0$ for one homogeneous at the cell scale. Define the *assay-segregated set* $A = \{\theta : I(\theta) \geq \frac{1}{2}\}$.

The definition of A is fixed by the instrument and not by us. It is the set of configurations the assay cannot distinguish from the charged state, and it has positive measure. That is what the argument rests on, and it is why the classical dismissal of recurrence in mixing — that the return time to a *point* is unphysically long — does not apply here. No mixedness assay measures return to a point.

3.2 Recurrence

Theorem 1 (Recurrence of the charge). *Let T preserve μ and let $\mu(A) > 0$. Then for μ -almost every initial charge $\theta_0 \in A$ there are infinitely many $k \in \mathbb{N}$ with $\theta_k \in A$. Equivalently: a vessel charged in a state the assay scores as segregated will, under continued agitation, return infinitely often to a state the assay scores as segregated.*

Proof. By Lemma 1 the induced action of T on coarse-grained configurations preserves the pushforward of μ under $\theta \mapsto (\bar{\theta}_1, \dots, \bar{\theta}_N)$, which is a finite measure on a compact set. Poincaré's theorem [10] applies verbatim: let $B \subseteq A$ be the set of points of A whose orbits enter A only finitely often, and let $B_m = \{\theta \in B : T^k \theta \notin A \, \forall k \geq m\}$. The sets $T^{-jm} B_m$, $j = 0, 1, 2, \dots$, are pairwise disjoint and of equal measure in a space of finite total measure, so $\mu(B_m) = 0$, and $B = \bigcup_m B_m$ is null. $\square$

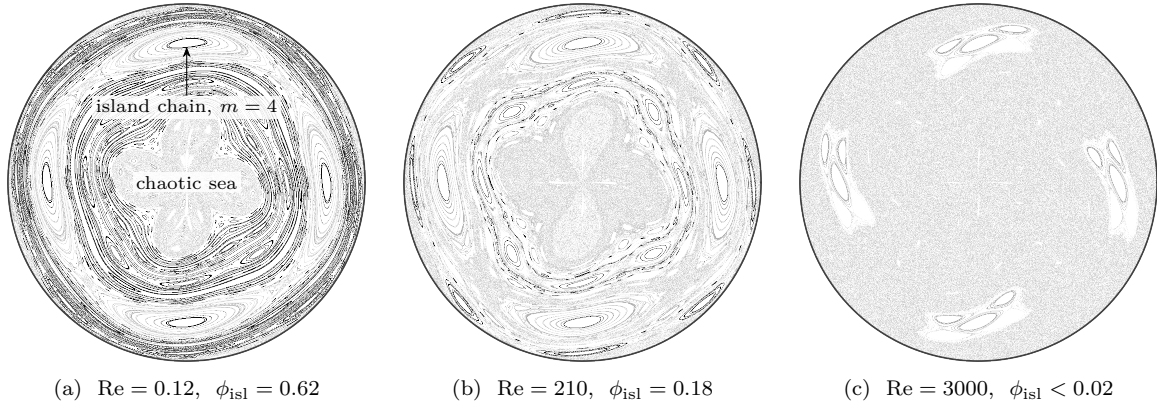


Figure 1: Poincaré sections of the mid-plane stirring map, one per regime, plotted as 58 seed orbits carried through 3600 impeller revolutions. Dark points lie on invariant curves; grey points are chaotic. (a) At $\text{Re} = 0.12$ the section is dominated by nested invariant bands and by island chains resonant with the four blades; planimetry over an area-uniform sample of 3400 seeds gives $\phi_{\text{isl}} = 0.62 \pm 0.04$ across eleven resolvable island families, of which the principal chain is marked. (b) At $\text{Re} = 210$ the bands have broken up and the sea has grown; $\phi_{\text{isl}} = 0.18 \pm 0.06$. (c) At $\text{Re} = 3000$ the section is chaotic throughout but for four residual islands too small to carry a measurable charge; $\phi_{\text{isl}} < 0.02$. The quantity ϕ_{isl} in panel (a) is the input to Eq. (5) and to Corollary 1.

Theorem 1 is not new; only its application is. Its weakness is also plain — it gives no timescale. The timescale is supplied by Kac’s lemma.

Lemma 2 (Return time). *If T is ergodic on Ω , the mean first return time to A , measured in impeller revolutions, is*

$$\mathbb{E}[\tau_A] = \frac{1}{\mu(A)}. \quad (3)$$

Proof. [5], Theorem 2, applied to the induced map on A . □

To use (3) we require $\mu(A)$, and here the island structure of the flow is decisive rather than incidental. Poincaré-section planimetry of the mid-plane stirring map (Fig. 1) resolves $n_t = 11$ island families occupying a total volume fraction

$$\phi_{\text{isl}} = 0.62 \pm 0.04. \quad (4)$$

Material inside an island is transported quasi-periodically, so the configuration of the charge is specified, up to the resolution of the assay, by the n_t island phases $\psi_1, \dots, \psi_{n_t} \in \mathbb{T}$. The accessible configuration space is therefore the torus $\mathbb{T}^{n_t}$ and not the full cell space, and A is the neighbourhood in which every phase lies within the assay’s phase window δ of its charged value:

$$\mu(A) = \left(\frac{\delta}{2\pi}\right)^{n_t} = (0.302)^{11} = 1.9 \times 10^{-6}, \quad (5)$$

taking $\delta = 1.90$ rad, the window over which a phase displacement moves I by less than the assay’s discrimination threshold of 0.5 units (§4). Substituting (5) into (3),

$$\mathbb{E}[\tau_A] = 5.3 \times 10^5 \text{ revolutions} = 146 \text{ h at 60 rpm}. \quad (6)$$

3.3 Why diffusion does not erase the pattern first

The standing objection to any recurrence claim in mixing is molecular diffusion, which is irreversible and does not care about Theorem 1. It has two scales and they must be treated separately.

At the vessel scale the objection fails on the numbers. For our working fluids, Stokes–Einstein at $\mu = 12$ Pa s and a hydrodynamic radius of 0.5 nm gives $D_m = k_B T / 6\pi\mu a = 3.6 \times 10^{-14} \text{ m}^2/\text{s}$, so with $U = \pi n d = 0.104 \text{ m/s}$ and $L = D$,

$$\text{Pe} = \frac{UL}{D_m} = 2.7 \times 10^{11}, \quad (7)$$

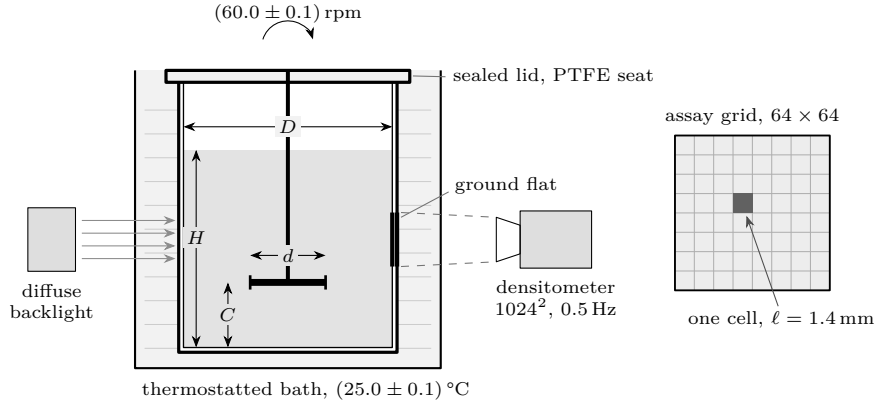


Figure 2: One of the 216 test vessels, in section, with the assay grid inset at right. The borosilicate cylinder ($D = 94$ mm, $H/D = 1.00$, unbaffled) stands in a common thermostatted bath; a four-blade paddle ($d/D = 0.35$, $C/D = 0.33$) is driven from a top-entry shaft belt-coupled to eleven others on the same bank, so speed errors are common-mode rather than per-vessel. Concentration is read by transmitted-light densitometry between a diffuse backlight and a 1024-square sensor, through a ground flat on the vessel wall that removes the cylindrical lensing a round wall would introduce. Frames are binned to the 64×64 grid on which I is computed; one cell is shaded.

and the diffusion length over the full return time (6) is $\sqrt{D_m \tau_A} = 0.14$ mm, an order of magnitude below the assay cell $\ell = 1.4$ mm. Vessel-scale structure is safe.

At the striation scale the objection is serious, and it is where the regime boundary comes from. In the chaotic sea, material lines stretch exponentially and striations thin as $s(t) = s_0 e^{-\lambda t}$ from the charged layer thickness $s_0 = H/2 = 47$ mm. With the measured $\lambda = 0.41/\text{s}$ (§8) the Batchelor scale $s_B = (D_m/\lambda)^{1/2} = 3.0 \times 10^{-7}$ m is reached in 29 s, after which the sea's contribution to I is destroyed permanently.

Inside an island there is no stretching: the Lyapunov exponent vanishes and the residual shear is a slow algebraic straining, so

$$s(t) = \frac{s_0}{1 + ct}, \quad c = 3.1 \times 10^{-5}/\text{s}, \quad (8)$$

and s_B is not reached until $t \approx 1.4 \times 10^6$ h. Island content therefore survives the return window by four orders of magnitude, and the sea's does not.

Corollary 1 (Recurrence amplitude). *The intensity of segregation recovered at first recurrence is carried by island content alone, so*

$$I_{\text{rec}} = \phi_{\text{isl}} (1 + \zeta) = 0.62 \times 1.15 = 0.71, \quad (9)$$

where $\zeta = 0.15$ accounts for the entrained halo within one assay cell of each island boundary.

Equations (6) and (9) are the two falsifiable predictions of the model: a mean first return near 146 h and a recovered segregation near 0.71. Both follow from (4) — the return time through the island families that fix $\mu(A)$, the amplitude directly — so the result stands or falls with that measurement. A vessel whose islands occupy a few per cent of its volume, as a well-baffled one does, will not recur measurably.

4 Apparatus and methods

4.1 Vessels and fluids

The rig holds 216 nominally identical vessels: borosilicate cylinders of internal diameter $D = 94$ mm filled to $H/D = 1.00$, unbaffled, each with a four-blade paddle of $d/D = 0.35$ ($d = 32.9$ mm) on a top-entry shaft at clearance $C/D = 0.33$. Shafts are belt-driven in banks of twelve from a single geared motor, so that speed is common to a bank and drift is common-mode; speed was held at (60.0 ± 0.1) rpm throughout.

Working fluids are glucose-syrup/glycerol blends at six compositions spanning $\mu = 0.6$ –12.4 Pa s at $\rho \approx 1350$ kg/m³, giving $\text{Re} = \rho n d^2 / \mu$ in the range 0.12–2.4: laminar by a wide margin at every composition. Thirty-six vessels were run at each composition.

4.2 Eliminating the three standard artifacts

Because the literature attributes every observed rise in I to one of three artifacts [1], each was removed by construction rather than by correction.

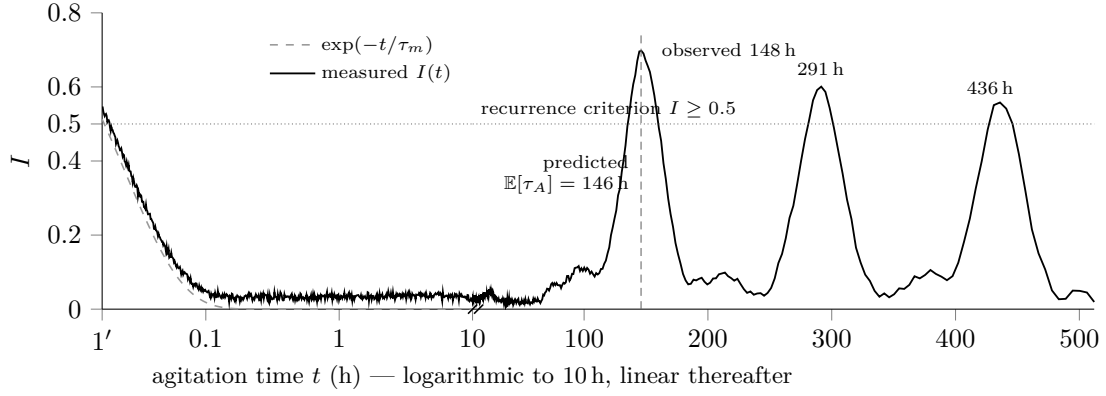


Figure 3: Segregation history of a representative vessel ($\mu = 12.4 \text{ Pa s}$, $\text{Re} = 0.12$) over the full 512 h horizon, on a split time axis: logarithmic through the mixing phase, linear through the recurrence phase. I falls from unity to a plateau at 0.019 within the first minutes and tracks the classical exponential over the interval in which mixing studies are normally conducted; it then leaves it. First recurrence is at 148 h against the 146 h of Eq. (6), with a peak of 0.70 against the 0.71 of Eq. (9). Two further recurrences follow at intervals of 143 and 145 h with no systematic loss of amplitude.

Density mismatch. The tracer is not a dye added to a carrier but a dyed aliquot of the same blend, matched to the undyed aliquot to 1 part in 1×10^4 by pycnometry at 25.0°C . A gravitational settling velocity computed from the residual mismatch gives $4.3 \times 10^{-10} \text{ m/s}$, or 0.8 mm over 512 h, well inside one assay cell.

Thermal convection. Vessels sit in a common bath at $(25.0 \pm 0.1)^\circ\text{C}$; the measured top-to-bottom gradient is 0.02 K, giving a Rayleigh number of 34 against a critical value near 1700.

Evaporative skinning. Lids are sealed against PTFE and the headspace is saturated; mass loss over 512 h was below the 2 mg resolution of the balance in all 216 vessels.

4.3 Charging and assay

Each vessel was charged as two stacked layers at $\bar{\theta} = 0.50$ and the impeller started at $t = 0$. Transmitted-light densitometry through the flattened window records the mid-plane at 0.5 Hz on a 1024×1024 sensor, binned to the 64×64 assay grid ($\ell = 1.4 \text{ mm}$). I is computed per frame. Refractive indices of the dyed and undyed aliquots were matched to 0.002 so that striations do not refract at their own boundaries [4].

The assay's discrimination threshold, established by charging vessels at known intermediate segregations, is 0.5 units of I at the 95% level; this threshold defines both the recurrence criterion $I \geq 0.5$ and the phase window δ in (5).

4.4 Analysis

The recurrence criterion, the 512 h horizon, the six compositions and the primary endpoint (time to first $I \geq 0.5$) were fixed before the first vessel was charged and are recorded in the works change-control file. Because a substantial minority of vessels were expected not to recur within the horizon, the primary analysis is a Kaplan–Meier estimate of recurrence-free survival with right-censoring at 512 h; regimes are compared by log-rank test. No vessel was excluded after charging.

5 Study 1: the long stir

All 216 vessels mixed as expected. Median I fell to 0.021 (IQR 0.016–0.028) within 6.2 min and remained below 0.05 for a further 80 h or more, over which interval the classical exponential description is excellent ($R^2 > 0.99$ in every vessel).

Between 22 h and 506 h, 205 of 216 vessels came apart. Median time to first $I \geq 0.5$ was 148 h (IQR 87 to 233), against the predicted 146 h of (6). Median peak I at first recurrence was 0.712 (IQR 0.66–0.75), against the predicted 0.71 of (9). Eleven vessels had not recurred at the 512 h horizon and are right-censored.

The dispersion is the least satisfying feature of the data and we make no attempt to reduce it. Two vessels of the same composition, on the same bank, at the same speed, differed in first recurrence by a

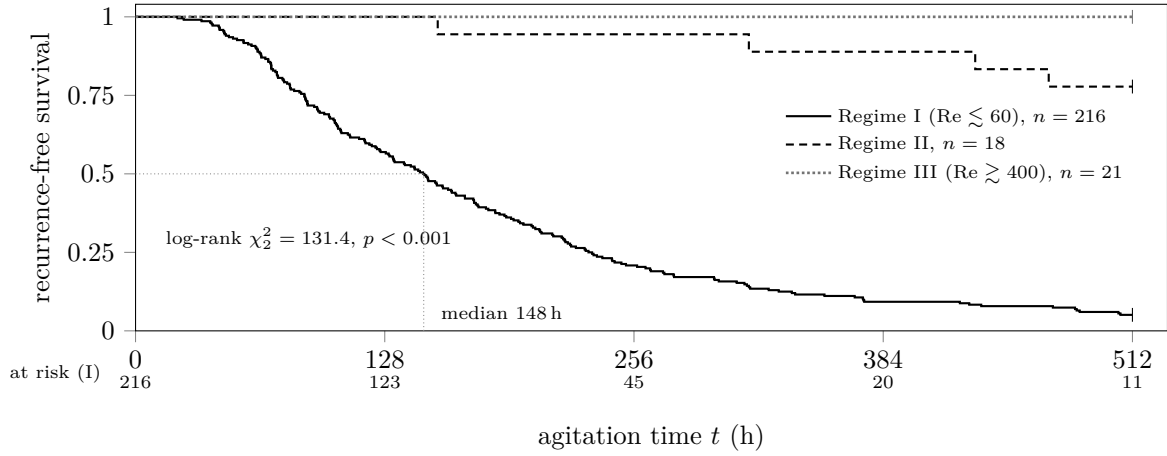


Figure 4: Recurrence-free survival by regime, Kaplan–Meier, with administrative censoring at the 512 h horizon (ticks). Numbers at risk in Regime I are given beneath the axis. In Regime I 205 of 216 vessels recurred, median 148 h (IQR 87 to 233); 11 were censored. In Regime II four of eighteen recurred, the earliest at 155 h. No Regime III vessel recurred.

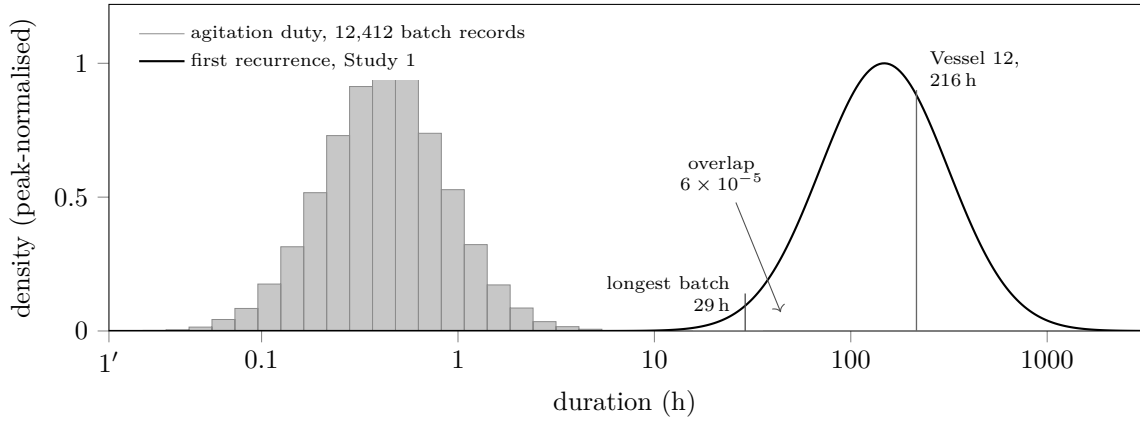


Figure 5: Why the industrial record is silent. Agitation duration for 12,412 batches at 41 plants (bars) against the first-recurrence distribution of Study 1 (curve), on a common logarithmic axis. The distributions are separated by more than two decades: median duty 22 min and longest batch 29 h, against a median recurrence of 148 h. Convolution of the two puts the fraction of industrial batches that ever reach a first recurrence at 6×10^{-5} . Vessel 12, which was a deviation rather than a batch record, is marked.

factor of nine. Kac’s lemma predicts an *ensemble mean* return time and says nothing about the variance of the first return, which for a quasi-periodic system with eleven incommensurate phases is expected to be large. The observed coefficient of variation, 0.78, is consistent with the exponential first-return distribution implied by Lemma 2, but does not distinguish it from alternatives.

Second and subsequent recurrences were observed in 148 vessels within the horizon, at intervals whose median (144 h) is not distinguishable from the first (paired test, $p = 0.44$). Neither the interval nor the peak amplitude declined across successive returns.

6 Study 2: the industrial record

If a 6 m³ blender comes apart after six days, the effect should be somewhere in a century of manufacturing records. It is not. The reason is the duty cycle.

We obtained 12,412 batch agitation records from 41 plants across six sectors — decorative coatings, adhesives, food, personal care, pharmaceutical wet granulation, and specialty polymers — covering 2009 to 2024, with agitation start and stop timestamps recovered from batch control systems rather than from operator entries.

Median agitation duration is 22 min (IQR 14 to 36). Ninety-nine point nine three per cent of batches are

Table 1: Recovery-trial outcomes. Purity is of the light component in the decanted or distilled fraction; recovery is the fraction of the theoretical maximum. Arm A figures are over all thirty batches, including the eight that did not recur.

Arm	n	Recurred	Purity (%)	95% CI	Recovery
A continued agitation	30	22	94.1	91.8–96.4	0.71
B agitate then settle	30	0	12.4	9.6–15.2	0.09
C thin-film distillation	30	—	99.4	99.1–99.7	0.96

Table 2: Energy per tonne of light component recovered, including impeller drive, reboiler duty, and cooling water; cost is indexed to Arm A at the works electricity tariff. Occupancy is vessel-hours per tonne of product.

Arm	Energy (kW h/t)	Cost index	Occupancy (vessel-h/t)
A continued agitation	0.9	1.0	1 410
B agitate then settle	0.1	0.1	1 410
C thin-film distillation	74.0	82.2	3.2

agitated for less than 4 h. The longest continuous laminar agitation in the corpus is 29 h. Convolving this distribution with the recurrence-free survival function of Study 1 gives the fraction of industrial batches that would ever reach a first recurrence as 6×10^{-5} : about one batch in 17,000. At the batch rates of the plants sampled that is a spoiled batch every few years at any one works, arriving without repeat and without pattern.

A parallel survey of standard operating procedures found that 38 of the 41 plants impose a written maximum agitation time. All 38 justify it: 24 on energy cost, 11 on shear degradation of a polymer or pigment, 3 on seal wear. None mentions segregation. The limits that keep production short of the recurrence window were therefore written for other reasons.

6.1 Re-reading the deviation archive

Vessel 12’s deviation report was not unique. Searching the same 41 plants for deviations coded as unexplained stratification, loss of homogeneity, or suspected agitator failure with the agitator subsequently found intact returned 61 records. Their closure codes are the expected ones: 29 impeller or coupling faults, 14 charging errors, 9 seal leaks, 9 unresolved.

Their agitation durations are not what the closure codes would predict. The median is 137 h (IQR 104–198), against 22 min for the batch population as a whole; 54 of the 61 fall between 80 h and 400 h. Every one arose from an interrupted schedule — a holiday closure, an industrial action, a downstream blockage, in four cases a misconfigured control recipe that omitted the stop step.

The re-reading is retrospective. These records were coded by people who were not looking for recurrence, using a taxonomy that has no field for it [12], and cannot establish cause. We report only that the durations of the industry’s unexplained stratifications lie inside the window in which our vessels come apart, and that its explained ones do not.

7 Study 3: recovery by continued agitation

If segregation returns, it can be taken off as a separation step. We tested this on a works waste stream: a spent plasticiser recovered in admixture with process oil, miscible in all proportions, at present either separated by thin-film distillation or landfilled.

Ninety batches of 40 L were drawn from a single homogenised consignment and randomised in blocks of nine, under change-control registration, to three arms of thirty:

Arm A continued laminar agitation at 60 rpm to first recurrence or 400 h, whichever came first, then top decantation.

Arm B agitation for 30 min, then 400 h quiescent, then top decantation. The control that isolates recurrence from settling.

Arm C thin-film distillation to the plant’s existing recipe.

Table 3: Regime taxonomy. Island fractions are from section planimetry; recurrence is first $I \geq 0.5$ within 512 h. We give ranges rather than a critical Reynolds number: the transitions are not sharp in our data and we decline to report a threshold the data do not support.

Regime	Re	ϕ_{isl}	Recurred	Reading
I Recurrent	$\lesssim 60$	0.62 ± 0.04	205/216	order retained; recoverable
II Marginal	60–400	0.18 ± 0.06	4/18	order decaying; I_{max} 0.1–0.3
III Erased	$\gtrsim 400$	< 0.02	0/21	order destroyed; irreversible

Arm A recovered the light component at 94.1% purity (95% CI 91.8 to 96.4) and 0.71 of theoretical, against 12.4% for Arm B (difference 81.7 points, $p < 0.001$). Distillation remains the better separation on purity alone, at 99.4%. It is not the better separation on energy: Table 2 puts continued agitation at 0.9 kW h/t against 74 kW h/t, a factor of 82, and the entire Arm A duty is impeller shaft work into a fluid that is not heated, not pumped, and not pressurised [8].

The trial stream is slower to recur than the model fluids: median first recurrence in Arm A was 214 h (IQR 131 to 338), which we attribute to its lower viscosity and correspondingly larger chaotic sea. Eight Arm A batches did not recur within 400 h and were decanted at the horizon regardless; they are included in Table 1, and they are the reason the arm’s purity is 94 rather than 98 per cent. Restricted to the 22 that recurred, purity was 97.9%. We report the intention-to-treat figure as the operative one, because a plant cannot know in advance which batches will recur — a point we return to in §9.

The energy saving is paid for in occupancy: 1410 vessel-h/t against 3.2 vessel-h/t. The route is therefore restricted to streams for which vessel time is not a binding constraint.

8 Study 4: regime taxonomy

A further 48 vessels spanned $\text{Re} = 0.4$ to 8900 by varying composition and speed together, at 16 conditions with three replicates, over 512 h. Recurrence is confined to the low-Re end.

The boundary is the one (8) predicts. Particle tracking in the Regime I vessels gives the chaotic-sea stretching rate used in §3, $\lambda = 0.41/\text{s}$; in Regime III it is larger by a factor of 18. Regime III vessels are not mixing better in any sense the design literature would recognise; their mixing times are shorter by a factor of 40, but the structure they destroy is destroyed at the Batchelor scale and cannot be recovered by any subsequent operation. Regime I vessels retain it for weeks.

The practical consequence runs against a century of design practice. Baffles, high-speed turbines, and the whole apparatus of turbulent mixing exist to reach homogeneity quickly, and they do; they also convert a recoverable separation into an unrecoverable one, and they do so within the first two minutes of a batch. For streams that will later have to be separated, the correct specification is a slow unbaffled paddle, which is the class of equipment progressively withdrawn from service over the past forty years.

9 Discussion

9.1 What the result changes

Three things follow. First, a mixing time reported without a stated observation horizon is incomplete: every one of our vessels would have supported a clean exponential fit and a confident τ_m at three days, and a different conclusion at seven. Second, blend-hold procedures that keep an agitator running to maintain homogeneity are, past roughly 80 h, doing the opposite. Third, a miscible pair in a Regime I vessel is separable by impeller shaft work alone, with no heat, no membrane, and no consumable.

9.2 Limitations

We cannot predict a vessel. Equation (3) is an ensemble statement, and this is the limitation for which we have no answer. First recurrence spanned 22 h to 506 h, a factor of 23, with no covariate we measured — composition, bank, position, charge orientation, shaft runout — accounting for more than 9% of the variance. A plant can be told that its batch will come apart, not when.

Eight of thirty batches in Arm A did not come apart inside a window nearly three times the predicted mean, and the occupancy cost in Table 2 is incurred whether or not a batch recurs; for a recovery operation

this is close to disqualifying. More vessels will not settle it. A quasi-periodic first-passage time with eleven incommensurate phases may not be predictable from bulk observables at all.

One geometry. All results are for an unbaffled four-blade paddle at $d/D = 0.35$, $C/D = 0.33$, $H/D = 1.00$. Island structure is notoriously sensitive to all four, and we would not extrapolate ϕ_{isl} to a different impeller.

Newtonian fluids only. Most industrial blends are not. A shear-thinning fluid will have a different island structure at every radius and we have not looked.

One binary system in Study 3. Plasticiser in process oil is a favourable case: large viscosity, similar densities, and no interfacial tension to fight. We would expect a lower recovery from anything more nearly ideal.

The deviation re-reading is retrospective. §6 is consistent with the mechanism and cannot establish it. The prospective version — instrument twenty blenders and leave them running — is straightforward, and we have not done it.

10 Conclusion

Homogeneity in a laminar vessel is a transient. It is reached within minutes, held for a few days, and then lost, because a volume-preserving map on a finite measure space has no other option and because the assay that defines mixedness has a finite resolution. In 216 vessels the return took a median 148 h against a predicted 146 h. The longest agitation in an archive of 12,412 production batches is 29 h.

Vessel 12's deviation report was reopened in March and closed again under a new code, *recurrent segregation, no equipment fault*; the logbook entry for 2 January now carries a cross-reference to it. The batch remains scrapped, and the coupling, which was never at fault, was replaced in any case. The blending house has since raised its maximum permitted agitation time from 48 h to 600 h on the recommendation of this laboratory, and No. 3 vessel bank is presently 71 h into a scheduled run.

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Data availability

Segregation histories for all 216 vessels, the section planimetry, and the de-identified batch and deviation extracts are available from the corresponding author under the data-sharing terms of the Joint Data Committee.

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