Spatially Optimized Multi-Shafts Stirred Reactors: An Experimental Study on Spatiotemporal Instabilities

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Abstract

Multi-shaft stirred reactors significantly impact the mixing efficiency and eliminate isolated mixed regions. Herein, the chaotic characteristics and spatiotemporal instability of flow fields were investigated in three different stirred reactor, and a novel flow field visualization technique was proposed to address limitations with low-viscosity fluids. Results demonstrated the chaotic behavior and energy transfer are inconsistent at various axial positions in the S-T-STR, D-T-STR, and T-T-STR flow fields, which are supported by pressure pulsation attractors, fractal dimension, largest Lyapunov exponents, multiscale entropy, and Kolmogorov entropy analysis. Torque signal attractor images confirmed stable periodic energy input to the flow field through the impeller. Hilbert spectrum analysis revealed time instability and periodic energy features after flow field stabilization. T-T-STR exhibited reduced spatial and temporal instability, denser periodic cycles, accelerated flow field structure evolution and energy transfer rate. These distinctions result from the distinctive flow field structure within each reactor configuration.

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Abstract

Multi-shaft stirred reactors significantly impact the mixing efficiency and eliminate isolated mixed regions. Herein, the chaotic characteristics and spatiotemporal instability of flow fields were investigated in three different stirred reactor, and a novel flow field visualization technique was proposed to address limitations with low-viscosity fluids. Results demonstrated the chaotic behavior and energy transfer are inconsistent at various axial positions in the S-T-STR, D-T-STR, and T-T-STR flow fields, which are supported by pressure pulsation attractors, fractal dimension, largest Lyapunov exponents, multiscale entropy, and Kolmogorov entropy.
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**Keywords:** multi-shafts stirred reactors; chaos; spatiotemporal instability; fractal dimension; flow field visualization.

**1 INTRODUCTION**

Stirred tank reactors (STRs) are operational units and key equipment in chemical processes, directly impacting the mass transfer and reaction efficiency, as well as process economics and the energy conservation\(^\text{1}\). Due to their high energy and economic efficiency, reliability, and ease of operation and maintenance, STRs are widely used in industries such as biotechnology\(^\text{2,4}\), food\(^\text{5}\), pharmaceuticals\(^\text{6,7}\), and metallurgy\(^\text{8}\). The large-scale and intelligent development of STRs is an inevitable trend in the advancement of chemical equipment\(^\text{9,10}\). To ensure that the ease of operation and manufacturability of STRs are not compromised during the scale-up process, high reliability of reactors is crucial\(^\text{11}\). Previous studies have shown that the height of the stirred tank required for coaxial stirring systems with three or more impellers is not applicable to industrial applications\(^\text{12}\). The scale-up of traditional chemical equipment does not always maintain similarity between laboratory and industrial scales and may even result in deterioration of mixing uniformity\(^\text{13}\). To meet the increased mixing demands in larger industrial volumes, the study of scaling-up mechanisms in STRs has highlighted the importance of
investigating the influence of vessel diameter on flow field structure and mixing performance\textsuperscript{14}. Petříček et al.\textsuperscript{15} proposed when the flow field characteristics are similar in stirred tanks of different diameters, the impact of scaling-up on overall mixing performance is minimal and may even have a positive effect on gas-liquid mass transfer coefficients. However, for the internal transport mechanisms and processes in the flow field of STRs, there are still scaling effects arising from spatial scale mismatch, which remains a critical scientific issue limiting the large-scale development of STRs. The complexities of mixing in STRs arise from the difficulties in physically analyzing the mechanisms and phenomena involved, contributed to the lack of reliable measurement devices. This has made scaling-up of mixing a direct challenge in many industrial processes, particularly in the amplification of STRs\textsuperscript{13}.

As a crucial component in STRs, the impeller transfers mechanical energy from electrical energy to the flow field, which influenced the formation, evolution, and transmission efficiency of the flow field. Research on traditional single-shaft, single-impeller STRs has revealed the tendency to form isolated mixed regions (IMRs) in the flow field\textsuperscript{16}. IMRs are coherent structures formed based on the free shear effect of the mixing layer\textsuperscript{17}. These regions exhibit high fluid rotational velocity and low mass exchange rate, resulting in energy consumption and reduced mixing efficiency\textsuperscript{18}. In fact, over 90\% of the energy in STRs is consumed to maintain the symmetric flow field structure formed by fluid rotation. In large-scale STRs, this phenomenon may be more prominent, as the spatial enlargement does not cause rapid instability of the flow field, but rather stabilizes it, leading to an increased energy consumption. As a result, more energy is required to overcome this flow field structure and create instability and chaos there in order to improve the mass transfer and mixing efficiency, especially in high
The most effective method to improve the mixing efficiency in stirred reactors is breaking the chaos isolation zone. Previous studies have proposed various methods to eliminate the IMRs, such as single shaft multi-impeller, different types of impeller combinations, single shaft eccentricity, dual-shaft eccentricity, and chaotic mixing. However, configurations with single shaft and multiple impellers still result in a stable and symmetric flow structure, only increasing the complexity of the flow field without fundamentally addressing the issue of mixing isolation zone. Studies have shown that eccentric multi-shaft configurations are more effective in breaking the chaos isolation zone and homogenizing the entire flow field, significantly reducing the mixing time. In addition, the use of eccentric configuration in combination with baffles in the reactor can greatly enhance the disruption of the symmetric flow structure, forming local vortices and increasing local mass transfer rates, resulting in a more complex flow field and further improving the mixing efficiency. Therefore, a multi-shaft eccentric overall configuration that permits free placement in the spatial domain is more suitable for large-scale mixing reactors. Moreover, for high-viscosity liquids, multi-shaft configurations are a better choice compared to the single shaft.

The number of impellers in a stirred reactor can impact the overall power consumption and mixing efficiency. Three impellers are the optimal number of configuration when considering comprehensiveness. In addition, the type of impeller also has a significant influence on the mixing efficiency and power consumption. Non-conventional shapes of impeller blades, such as grooved blades, fractal blades, and combinations of rigid and flexible blades, can be used to eliminate the chaos isolation zone and improve mixing.
efficiency and can be optimized using multi-objective genetic algorithms (MOGA)\textsuperscript{31}. However, studies have shown that conventional radial blades are more suitable for macro-scale mixing in large-volume reactors\textsuperscript{32}. Luan et al.\textsuperscript{33} indicated that the chaotic characteristics decrease with the increasing rotational speed in the single-shaft flow field. However, maintaining a high degree of chaos and disorder in the internal flow field of a stirred reactor is practically meaningful as it leads to higher mass transfer rates and further enhances the macro-scale mixing efficiency. These studies provide a good description of the flow field in different regions of the mixing vessel, which helps in determining the performance and mixing efficiency of the stirring mixer\textsuperscript{34}. Moreover, the interaction between the impeller jet and the reactor wall can result in periodic energy fluctuations\textsuperscript{35}.

Capturing the dynamics and disorderliness of the flow field changes in stirred reactors is relatively challenging. Currently, the most convenient and effective non-intrusive visualization techniques involve acid-base visualizations and ultraviolet (UV) fluorescence techniques\textsuperscript{17}. Previous studies have shown that these techniques can accurately determine the mixing residence time and chaotic segregation zones\textsuperscript{20,37}, and UV fluorescence techniques can be used to identify the critical Reynolds number for disrupting chaotic segregation zones with different impeller structures\textsuperscript{25}. Meanwhile, researchers have also explored the generation mechanism and internal three-dimensional structure of chaotic segregation zones and proposed that enhanced axial circulation is the key to eliminating chaotic segregation zones\textsuperscript{17}. In recent years, new methods such as luminescent capsules\textsuperscript{37} and ultrasonic sensors\textsuperscript{38} have emerged for characterizing chaotic segregation zones and mixing time, but these methods are based on experiments with single-shaft, single-impeller stirred reactors. Currently, no relevant research
has been found for visualizing flow field structures in large-diameter multi-shafts stirred reactors, which is highly meaningful for the research of scale-up in stirred reactors. Flow field visualization techniques are valuable in identifying flow field instabilities and transition points between flow regimes, which are crucial for industrial applications and further in-depth investigations.

Previous studies have mainly focused on the relationship between flow field instabilities, such as chaos and flow velocity. However, there has been relatively little research on the treatment details of easily measurable passive scalars in the flow fields of agitated reactors. Nonetheless, the changes in flow field structure in agitated reactors can give rise to highly nonlinear dynamic characteristics in various physical quantities, which hold significant importance in turbulence research\(^39\). Previous studies have found local instabilities in agitated reactors and observed the existence of these instabilities in the overall chaotic characteristics of the flow field\(^40\). Fluid agitation is a complex nonlinear process, and the irregularity of fluid motion and multiscale behavior during mixing inevitably leads to chaotic phenomena in the flow field, with local chaotic phenomena being more prominent in scaled-up reactors and multi-shaft agitated reactors.

In this work, we used glycerol-water solutions of varying viscosities in three different configurations of stirred reactors. The evolution of flow field structures was analyzed using fluorescent flow field visualization techniques and a novel approach that combines flow field visualization with image processing was proposed to overcome the limitations of fluorescent visualization for low-viscosity fluids. Subsequently, we performed macroscopic comparisons of parameters such as power consumption, maximum Lyapunov exponent, multiscale entropy,
and Kolmogorov entropy to evaluate the overall chaotic behavior and energy transfer in the flow field and combined with the attractor image, the instability characterization of energy and local spatial chaos characteristics is obtained. Furthermore, by applying the Hilbert-Huang transform, macroscopic and local temporal instability features were extracted from nonlinear physical quantities. This study highlights the advantages of multi-shafts stirred reactors in terms of the scalability and intelligent promotion, providing a comprehensive understanding of flow field structure evolution and spatiotemporal instability characteristics.

2 EXPERIMENTAL SECTIONS

2.1 STR configuration

In this study, we used organic glass cylindrical stirred tank reactors with three different configurations: a single-shaft three-impeller reactor (S-T-STR), a double-shaft three-impeller reactor (D-T-STR), and a triple-shaft three-impeller reactor (T-T-STR) (Figure 1). The reactors had aspect ratios of 2:1, 1:1, and 1:2, respectively, with a working volume of 20 L each. Baffles were installed in all reactors, with a width of 1/10 of the reactor diameter and a thickness of 3 mm. The impellers used were traditional six-bladed axial impellers, with constant diameter. The minimum distance between the impeller and the bottom of the tank was set at 1/6 of the liquid level height, and the distance between adjacent impeller was set at 1/4 of the liquid level height. Details of the impeller can be found in Supplementary Figure 1 and the specific parameters of the reactors and impeller in Supplementary Tables 1 and 2.
FIGURE 1 Experimental equipment: (A) schematics of experimental setup for T-T-STR, (B) actual picture of experimental device, (C) geometry of S-T-STR, (D) geometry of D-T-STR.

2.2 Materials and experimental apparatus

The chemicals used were as follows: glycerol (glycerin, mole fraction purity 0.990), sodium fluorescein (uranine), pearlescent powder (1000 mesh), sodium hydroxide solution (NaOH, 0.001 mol/m³), and hydrochloric acid (HCl, 0.001 mol/m³). All chemicals in the experiments
were purchased from Shanghai Macklin Biochemical Co., Ltd. without further purification. The experimental media consisted of mixtures of water and glycerol with varying volume ratios, and their properties were characterized at each ratio, as summarized in Supplementary Table 3. The viscosity ($\mu$) and density ($\rho$) of working medium were measured by digital viscometer (Shanghai Jingtian Electronic Instrument Co., Ltd., SNB-2) and pycnometer, respectively. The experimental apparatus consisted of the following parts: motor (Shaoxing Wanpeng Electromechanical Co., Ltd., YSB7104), high frequency dynamic pressure sensor (Nanjing Aier Sensing Technology Co., Ltd., AE-S), dynamic torque sensor (Sanhe Yanjiao Huaxin Electromechanical Co., Ltd., HX-901), LED professional film and television lights (Jinbei., EFIII-200), high speed camera (Revealer., 5F04M), UV purple light (LG., 800W-405nm), and camera (Canon., 80D).

2.3 Experimental procedures

2.3.1 Flow Field Visualization

We utilized the strong fluorescence property of sodium fluorescein under alkaline conditions in this study. A basic solution was prepared by adding NaOH and glycerol-water solution, which was then poured into containers to a predetermined water level. After achieving a certain impeller speed and sufficient time, a small amount of acidic solution (HCl) was injected near the sidewall at half depth, causing the green dye to decolorize and form IMRs through neutralization reactions. A fluorescent tracer was subsequently injected into the IMR region using an injection pump to visualize its three-dimensional morphology. However, this method is limited to higher viscosity fluids.

To overcome this limitation, we innovatively used phosphorescent powder as a flow field
tracer. The same mass of phosphorescent powder was added to different systems and configurations, uniformly mixed, and illuminated using an LED light for supplementary lighting. The impeller speed was adjusted, and flow field images were captured using a camera and high-speed camera at 1000 FPS. The differential box counting method was employed to extract the two-dimensional fractal dimension of the flow field images at different rotational speeds. The fractal dimensions were calculated separately for the two camera positions and then averaged to obtain the final fractal dimension, as shown in Figure 2.

![Fractal dimension image extraction for (A) camera and (B) high-speed camera.](image)

F I G U R E 2 Fractal dimension image extraction for (A) camera and (B) high-speed camera.

The two-dimensional fractal dimension is a measure used to characterize and describe the complexity of complex shapes. It not only quantifies the complexity of an image, but also exhibits invariance to multiscale and multiresolution changes. This concept can be effectively applied to assessing the chaotic nature of flow fields. Chaudhuri and Sarkar\textsuperscript{42} proposed the differential box-counting (DBC) method for calculating the fractal dimension. The fractal dimension $D$ can be estimated by the least squares fitting of $\log(N_r)$ and $\log(1/r)$:

$$y = Dx + c$$

(1)
where \( y = \log(N_r), \) \( x = \log(1/r), \) \( r \) is partition ratio and \( N_r \) the total number of boxes in all grids.

2.3.2 Pressure pulsation signal extraction and processing

The wall pressure fluctuations on the stirrer tank are monitored by sampling at a frequency of 1000Hz at different axial positions, rotational speeds, working fluids, and structural configurations. The pressure fluctuation signals are then processed as follows:

(1) Chaotic attractor

The morphology and structure of the chaotic attractor in phase space are important criteria for evaluating the characteristics of turbulent flow\(^{43}\). Assuming that any transient pressure fluctuation signal is represented by \( p_t = \{ p_t(i), i=1,2,\cdots,n \} \), where \( n \) is the total length of the data sequence and its normalized time series is represented by \( p(i) \). The delay reconstruction method is used to reconstruct the phase space by selecting an appropriate embedding dimension \( m \) and delay time \( \tau \) to embed the single-variable into the \( m \)-dimensional Euclidean space \( \mathbb{R}^m \). The phase space element is denoted as:

\[
P_i(m, \tau) = [ p_k, p_{k+\tau}, p_{k+2\tau}, \ldots, p_{k+(m-1)\tau} ]
\]  

(2)

where \( k=1,2,\cdots,N \), and \( N \) is the total number of points on the reconstructed phase space attractor, \( N=n-(m-1)\tau/\Delta t \), where \( \Delta t \) is the sampling time interval. The “Delay Coordinate Embedding” and “Mutual Information” are used to determine the optimal delay time and embedding dimension\(^{44,45}\).

(2) Largest Lyapunov exponents (LLE)

The Lyapunov exponent is a key parameter in characterizing the dynamics of a system by measuring the convergence or divergence rate between neighboring trajectories in phase space\(^{46}\). It plays a significant role in determining the chaotic behavior of a nonlinear time
The approach introduced by Wolf et al. involves reconstructing the phase space from the evolution of phase trajectories, planes, and volumes, enabling the extraction of the Lyapunov exponent from a single-variable time series.

Starting from an initial point \( Y(t_0) \) and its nearest neighbor \( Y_0(t_0) \) with a distance of \( l_0 \), the time evolution of these two points is tracked until their separation distance exceeds a preset value \( \delta > 0 \), denoted by \( l' = |Y(t_1) - Y_0(t_1)| > \delta \). At this point, \( Y(t_1) \) is kept and a new point \( Y_1(t_1) \) is found nearby with a distance of \( l_1 = |Y(t_1) - Y_0(t_1)| < \delta \) and as small an angle as possible. This process is repeated until \( Y(t) \) reaches the end of the time series at \( N \), with a total number of iterations denoted by \( M \). The LLE is then calculated as the limit of the average logarithmic separation rate of all pairs of nearby trajectories, which is given by the formula:

\[
\sigma_1 = \frac{1}{t_M - t_0} \sum_{i=0}^{M-1} \ln \frac{l_{i+1}}{l_i}
\]

(3) Multiscale entropy (MSE)

Multiscale entropy is a method to compute the sample entropy of a time series at multiple scales, reflecting the irregularity of the time series at different scales and providing better resistance to noise and interference, making it a more systematic approach for time series analysis.

Multiscale entropy involves three parameters: \( \varepsilon, m, \) and \( r \). \( \varepsilon \) is the scale factor, \( m \) is the embedding dimension, and \( r \) is the threshold. For a given scale factor \( \varepsilon \), the time series is transformed into a new series of length \( I=n/\varepsilon \) using coarse graining. \( C_{\varepsilon,m}^{i}(r) \) is defined as the ratio of \( M_i \) to \( I-m+i \), where \( i=1, 2, \ldots, I-m+1 \), and \( i \neq j \). Multiscale entropy is the set of sample entropy at multiple scales, where \( \varepsilon=1~15 \). Multiscale entropy can be expressed as:

\[
MSE = \left\{ \varepsilon \left| SampEn(\varepsilon, m, r) = \ln \left[ \frac{C_{\varepsilon,m}^{i}(r)}{C_{\varepsilon,m}^{i+1}(r)} \right] \right. \right\}
\]

(4)
(4) Kolmogorov entropy ($K_2$)

Using the joint probability of trajectories falling into boxes at different time intervals based on Shannon theory, the Kolmogorov entropy can be calculated. Therefore, extracting the Kolmogorov entropy directly from the phase space trajectory is an important problem for researchers. To address this issue, P. Grassber and I. Procaccia proposed an estimation method for the correlation entropy $K_2$ based on $q$-order Renyi entropy, with $q=2$. The $K_2$ can be obtained:

$$K_2 = \lim_{\Delta t \to 0} \lim_{n \to \infty} \lim_{t_n \to \infty} \frac{1}{n\Delta t} \log \left( \sum_{i \in L} P^{2}_{i \in L} \right)$$ (5)

(5) Hilbert-Huang Transform (HHT)

Norden E. Huang et al. introduced the HHT, a novel signal time-frequency analysis theory. The signal is accurately represented as a distribution of frequency-time-energy, known as the Hilbert spectrum. For any time series $x(t)$ decomposed by Empirical Mode Decomposition (EMD). Taking time and frequency as independent variables and amplitude as dependent variable, Hilbert amplitude spectrum can be obtained:

$$H(f, t) = \text{Re} \sum_{j=1}^{n} a_j(t) e^{i f_j(t) dr}$$ (6)

where $a_j(t)$ is the instantaneous amplitude function of the signal, $\theta_j(t)$ is the instantaneous phase function of the signal, and $f_j(t)$ is the instantaneous frequency function of the signal.

2.3.3 Torque pulsation signal extraction and unit volume power consumption processing

The torque signals of stirred reactors with different structural configurations, operating at varying viscosity of the working fluid and different speeds, are measured and stored using dynamic torque sensors. The power consumption per unit volume ($P_v$) is calculated using the
The torque fluctuation signals can be subjected to the same processing procedures as pressure fluctuation signals. Chaotic characteristic signals are extracted from the torque fluctuations and compared with the chaotic characteristics of pressure fluctuations. This comparison allows for a true assessment of the input of chaotic characteristics and their manifestation in the flow field, revealing the regularity of energy dissipation.

3 RESULTS AND DISCUSSION

3.1 Analysis of flow field visualization

F I G U R E 3 Flow field fluorescence visualization experiment for (A) S-T-STR with 90 \% glycerol solution as working fluid at the second impeller, (B) S-T-STR with 90 \% glycerol solution as working fluid between the second and third impellers, (C) S-T-STR with 50 \% glycerol solution as working fluid, (D) D-T-STR with 50 \% glycerol solution as working fluid and (E) T-T-STR with 50 \% glycerol solution as working fluid.
A flow field fluorescence visualization experiment was conducted in the S-T-STR using a 90% glycerol-water solution at a constant rotational speed of 60 RPM. The results, shown in Figure 4A and B, reveal the presence of chaotic segregation zones above and below the impeller. These zones tend to stabilize into a consistent internal circulation structure as the rotational speed remains constant. The interaction between the impellers creates a distinct interface. However, increasing the rotational speed does not improve substance transfer between the impellers. Subsequently, another experiment was performed in the S-T-STR using a 50% glycerol-water solution. At a rotational speed of 60 RPM, no significant chaotic segregation zones are observed, although the interface effect between the impellers is still present but less prominent. Increasing the mixing time and rotational speed leads to the disappearance of both the interface effect and chaotic segregation zones (Figure 3C). In the D-T-STR and T-T-STR, flow field fluorescence visualization experiments were conducted using a 50% glycerol-water solution at a rotational speed of 60 RPM. Notably, no significant chaotic segregation zones or interface phenomena are observed. However, both systems exhibit a vortex region near the impeller interaction area, promoting substance dispersion (Figures 3D and E).

3.2 Fractal dimension analysis of flow field structure

Figure 4 demonstrates the two-dimensional fractal dimension of the S-T-STR, D-T-STR, and T-T-STR flow fields. It is observed that the fractal dimension of the flow field structure in S-T-STR exhibits pronounced and periodic oscillations with varying rotational speeds at different axial positions. The difference between the maximum and minimum values of the fractal dimension at different axial positions ranges from 4.54% to 10.62%. Furthermore, there is a significant variation in the fractal dimension at different heights, with differences ranging from...
0.91% to 2.04%, and an average difference of 1.47%. In the case of D-T-STR, as the rotational speed increases, the periodic oscillations diminish, and the structure of flow field tends to become more homogeneous. The difference between the maximum and minimum values of the fractal dimension at different heights ranges from 3.57% to 8.09%. The disparity in the fractal dimension at different heights ranges from 0.23% to 2.34%, with an average difference of 0.91%. Compared to S-T-STR, D-T-STR exhibits smaller oscillations at different positions with increasing rotational speed. In T-T-STR, the periodic oscillations at different axial positions are relatively weak. The difference between the maximum and minimum values of the fractal dimension at different axial positions ranges from 2.62% to 5.10%, which is significantly smaller than that observed in S-T-STR and D-T-STR. Moreover, there is relatively little disparity between the fractal dimensions at different axial positions, with differences in the fractal dimension at different heights ranging from 0.45% to 1.81%, and an average difference of 0.72%. This indicates that the flow field structure in T-T-STR is not highly sensitive to axial positions and exhibits overall uniformity, which facilitates efficient substance transfer.

3.3 Flow field energy input analysis

Figure 5 investigates the influence of rotational speed on the $P_V$ in S-T-STR, D-T-STR, and T-T-STR.
T-STR for different viscosity working media. It can be observed that the $P_V$ of the six shafts increases with increasing rotational speed. Due to the fact that S-T-STR has three impellers mounted on a single shaft, resulting in a larger interaction area between the impellers and the fluid, the $P_V$ for rotational speeds above 200 RPM is significantly higher compared to the other five shafts in D-T-STR and T-T-STR for different viscosity working media. An interesting phenomenon is that the $P_V$ of D-T-STR-Single impeller is higher than that of D-T-STR-Double impeller. This phenomenon is also observed in T-T-STR, where the $P_V$ of T-T-STR-Middle is significantly higher than that of T-T-STR-Low and T-T-STR-High. This indicates that when the impellers are eccentrically placed at different axial positions in the mixing tank, there is interaction between the impellers at different axial positions through the fluid. Moreover, for structures with multiple eccentric shafts, the impeller at the middle position in the mixing tank is simultaneously influenced by the upper and lower impellers.

![Figure 5](image)

**Figure 5** Analysis of power consumption per unit volume for (A) the viscosity of the working medium is 1.26, (B) the viscosity of the working medium is 3.79 and (C) the viscosity of the working medium is 12.30.

Table 1 presents the average $P_V$ under different viscosity working media. As the viscosity increases, the $P_V$ of S-T-STR exhibits a decreasing trend, with an overall decrease of 14.08%. The $P_V$ of D-T-STR-Single impeller and D-T-STR-Double impeller increases with increasing viscosity, with respective increases of 8.63% and 14.52%. T-T-STR-Low, T-T-STR-Middle,
and T-T-STR-High exhibit an overall decrease in $P_V$ of 9.20%, 12.40%, and 9.22%, respectively, with increasing viscosity.

### TABLE 1 Average power consumption per unit volume under different viscosity working medium

<table>
<thead>
<tr>
<th>Specification</th>
<th>Stirring shaft</th>
<th>Viscosity=1.26 mPa • s</th>
<th>Viscosity=3.79 mPa • s</th>
<th>Viscosity=12.30 mPa • s</th>
</tr>
</thead>
<tbody>
<tr>
<td>S-T-STR</td>
<td></td>
<td>40.23 (W/m)$^3$</td>
<td>35.15 (W/m)$^3$</td>
<td>34.57 (W/m)$^3$</td>
</tr>
<tr>
<td>D-T-STR</td>
<td>D-T-STR-Single impeller</td>
<td>27.43</td>
<td>27.76</td>
<td>29.88</td>
</tr>
<tr>
<td></td>
<td>D-T-STR-Double impeller</td>
<td>17.84</td>
<td>20.32</td>
<td>20.43</td>
</tr>
<tr>
<td></td>
<td>T-T-STR-Low</td>
<td>20.65</td>
<td>19.09</td>
<td>18.75</td>
</tr>
<tr>
<td></td>
<td>T-T-STR-Middle</td>
<td>65.65</td>
<td>61.28</td>
<td>57.51</td>
</tr>
<tr>
<td></td>
<td>T-T-STR-High</td>
<td>20.50</td>
<td>19.34</td>
<td>18.61</td>
</tr>
</tbody>
</table>

Figure 6 illustrates the attractor plots of torque signals for S-T-STR, D-T-STR-Single impeller, and T-T-STR-Middle at different rotational speeds. Increasing rotational speed results in a linear growth trend in the attractor plots for all three systems. S-T-STR exhibits a two-dimensional limit cycle surface, while D-T-STR-Single impeller and T-T-STR-Middle display limit cycle attractor states with varying numbers of limit cycles. As the rotational speed increases, more limit cycles appear, indicating stable periodic energy input. In D-T-STR-Single impeller, the number of limit cycles ranges from 1 to 3 and potentially extends to 4, while in T-T-STR-Middle, it increases from 2 to 4 in a sequential top-to-bottom manner. The average power consumption per unit volume is higher in T-T-STR-Middle compared to D-T-STR-Single, indicating a relationship between the number and magnitude of torque limit cycles.

Varying viscosities have minimal impact on the attractor plots' morphology, primarily affecting the mapping area. The torque attractor is not significantly influenced by the working medium's viscosity, as demonstrated in Supplementary Figure 2.
FIGURE 6 Torque attractor at different RPM for (A) S-T-STR, (B) D-T-STR-Single impeller and (C) T-T-STR-Middle.

3.4 Time instability analysis

In the analysis of torque attractors, it was found that energy enters the flow field through the interaction between the impellers and the fluid in the form of periodic pulses. After energy enters the flow field, it evolves through the flow field structure, forming vortical structures of different scales. There is a periodic cycle of "appearance-dissipation-reappearance" of these vortical structures. The energy transfer within this process influences the variations of physical quantities in a wave-like manner. Therefore, analyzing these physical quantities can indirectly reflect the energy transfer process and the evolution of flow field structures.

Figure 7 to 9 shows the Hilbert spectrum of the pressure fluctuation collected at position \( P_3 \) in S-T-STR, D-T-STR and T-T-STR. It can be observed that the Hilbert spectrum exhibits fluctuations in energy over time, indicating significant temporal instability. These fluctuations become more pronounced with increasing rotational speed. In terms of peak energy values, the number of energy peaks in S-T-STR increases from 5 to 7 as the rotational speed ranges from 30 to 300 RPM. However, the corresponding frequencies of these peaks decrease with increasing rotational speed.

In low viscosity stirred reactors, turbulent motion dominates the flow field, characterized by the superposition of vortices at different scales. Large-scale vortices, influenced by...
boundary conditions and inertia, contribute to low-frequency fluctuations and can be comparable in size to the flow field. Conversely, small-scale vortices, determined by viscosity, have sizes approximately one-thousandth that of the flow field and are responsible for high-frequency fluctuations. Peaks in the Hilbert spectrum represent the continuous alternation of vortices at different scales, marking the end of an evolution cycle. This alternation involves the dissipation of small-scale vortices and the emergence of large-scale vortices.

At low rotational speeds, just before the occurrence of peaks, there is a sharp increase in the energy peaks located between high and low frequencies. However, this phenomenon is not observed at later time intervals after the peaks. This indicates that just before the dissipation of small-scale vortices, intermediate-scale vortices experience a rapid increase, suggesting that intermediate-scale vortices act as a bridge for energy transfer between large-scale and small-scale vortices. To enhance energy transfer and accelerate the evolution of flow field structures, the generation rate of intermediate-scale vortices needs to be increased. At high rotational speeds, there is an increased number of peaks within the same time interval, indicating an accelerated energy transfer and flow field structure evolution. This can be observed from the corresponding frequencies of the Hilbert spectra for high and low rotational speeds.

In comparison to S-T-STR, where the peaks appear in a pointed form, the peaks in D-T-STR appear in a concentrated manner within local regions. However, they also exhibit significant temporal instability. This is due to the eccentricity of the stirring shafts, which intensifies the interaction between the impellers and leads to the formation of a larger vortex region within the flow field. This accelerates the energy transfer between vortices of different scales and promotes the evolution of flow field structures. As a result, the vortex evolution is
no longer solely driven by the flow inertia. This observation is consistent with the findings from previous flow field fluorescence visualization experiments.

![Hilbert spectrum of pressure pulsation at $P_3$ of S-T-STR for (A) 30RPM, (B) 120RPM, (C) 210RPM and (D) 300RPM.](image)

**FIGURE 7** Hilbert spectrum of pressure pulsation at $P_3$ of S-T-STR for (A) 30RPM, (B) 120RPM, (C) 210RPM and (D) 300RPM.

In the Hilbert spectrum of D-T-STR, each peak region contains 1 to 3 peaks. This is only observed in high-speed conditions for S-T-STR. The presence of multi-peaks within the peak regions of D-T-STR is already evident at 30 RPM. As the rotational speed increases, the number of peak regions decreases, and the characteristics of single-peaked regions become more prominent. This indicates that increasing the rotational speed enhances the interaction between the impellers, concentrating the energy transfer within the vortex region generated by the interaction. This vortex region gradually dominates the entire flow field. Due to the increased interaction between the impellers, energy loss is intensified, resulting in a decrease in the energy transfer rate and the rate of flow field evolution.

Continuing to increase the rotational speed leads to a slow increase in the number of peak regions. When the speed reaches 300 RPM, a pattern emerges where multi-peaked and single-
peaked regions alternate within the peak regions. This suggests that as the rotational speed increases, the vortex regions generated by the interaction between the impellers no longer dominate the flow field for extended periods. Instead, the individual influence of each impeller on the flow field becomes more pronounced.

Figure 8 Hilbert spectrum of pressure pulsation at $P_3$ of D-T-STR for (A) 30RPM, (B) 120RPM, (C) 210RPM and (D) 300RPM.

In contrast to the alternating single-peak and multi-peak characteristics observed in the Hilbert spectra of S-T-STR and D-T-STR, T-T-STR exhibits a dense arrangement of single peaks. As the rotational speed increases, the number of single peaks in T-T-STR increases, and their arrangement becomes even denser. This indicates that the interaction between the impellers in the T-T-STR flow field does not hinder or disrupt the energy transfer within the flow field. Instead, it promotes the rate of energy transfer and the evolution of flow field structures. Furthermore, the small frequency span in T-T-STR suggests that the scale changes within the flow field occur smoothly and rapidly. The energy transfer rate within the flow field remains consistently high and stable. This observation is also supported by flow field
fluorescence visualization experiments, where the interaction regions within the flow field enhance the overall evolution rate and energy transfer rate, leading to a more chaotic flow field.

Overall, considering the same working fluid and rotational speed conditions, the T-T-STR exhibits superior flow field evolution and energy transfer rates compared to S-T-STR and D-T-STR. Additionally, it demonstrates the least temporal instability.

![Hilbert spectrum of pressure pulsation at $P_3$ of T-T-STR for (A) 30RPM, (B) 120RPM, (C) 210RPM and (D) 300RPM.](image)

3.2 Spatial instability analysis

Figure 10 to 12 illustrates the chaotic attractors corresponding to pressure fluctuations in S-T-STR, D-T-STR and T-T-STR at different axial positions, fluid viscosities, and rotational speeds. It is evident that the shape of the chaotic attractor in S-T-STR shows no significant correlation with axial position or rotational speed. On the other hand, the fluid viscosity affects the enclosed area of the attractor phase plane. Higher viscosity results in a smaller enclosed area. This is because increased viscosity leads to a more stable interface effect, hindering the energy transfer process.
The chaotic attractor provides insights into the system's developmental trends, and its morphology partially reflects the degree of chaos within the system. The level of chaos directly determines the flow field's ability to disperse matter. In the case of S-T-STR, the flow field's chaotic degree cannot be significantly enhanced by varying the rotational speed. Additionally, there are no unstable regions in its spatial domain, limiting the utilization of phase differences to enhance the chaotic characteristics of flow field.

For D-T-STR, both axial position, viscosity, and rotational speed have significant impacts on the shape and area of the chaotic attractor. At lower axial positions, moderate fluid viscosities, and lower rotational speeds, a double-vortex chaotic attractor morphology is observed. However, this morphology is not stable as axial position, rotational speed, and viscosity vary, exhibiting characteristics of metastable structure. Although no periodic features are observed, it indicates that increasing the energy input path, such as increasing the number of impellers, is feasible for enhancing the chaotic degree of flow field. Therefore, within D-T-STR, spatial instability exists, implying the presence of certain phase differences between different axial positions, which can accelerate energy transfer and flow field evolution in space. Moreover, the overall chaotic degree of the D-T-STR flow field can be controlled by adjusting the rotational speed, thereby improving flow field mixing and dispersion performance.

FIGURE 10 Pressure pulsation attractor of S-T-STR at (A) different axial locations, (B) different viscosity working medium and (C) different RPM.
FIGURE 11 Pressure pulsation attractor of D-T-STR at (A) different axial locations, (B) different viscosity working medium and (C) different RPM.

FIGURE 12 Pressure pulsation attractor of T-T-STR at (A) different axial locations, (B) different viscosity working medium and (C) different RPM.

The axial position had a minimal influence on the morphology of the chaotic attractor in T-T-STR. However, it had a significant impact on the position in phase space, indicating substantial phase differences between axial positions, resulting in a fast and uniform energy transfer rate and a consistent overall chaotic degree of the flow field. On the other hand, the morphology and area of the attractor show minimal sensitivity to different fluid viscosities. With increasing rotational speed, the morphology of the attractor does not undergo significant changes, but its area tends to decrease to some extent. This suggested the presence of a flow field structure that requires energy input to sustain, namely, the interaction between impellers at different axial positions. However, unlike the interaction vortical regions observed in D-T-STR, this flow field structure in T-T-STR does not hinder energy transfer but rather possesses the capability to enhance and balance the overall chaotic degree of the flow field.
Figure 13 illustrates the variation of LLE with rotational speed for S-T-STR, D-T-STR, and T-T-STR at different axial positions. S-T-STR, D-T-STR, and T-T-STR exhibit different degrees of fluctuation in LLE with changing rotational speed. Additionally, there are numerical differences in LLE values among different axial positions, indicating spatial instability in the chaotic degree of the flow field. Both S-T-STR and D-T-STR show a decrease in LLE with increasing rotational speed, with S-T-STR exhibiting a larger decrease compared to D-T-STR. The average LLE values for the same axial positions but different rotational speeds are approximately 0.38-0.47, 0.25-0.34, and 0.57-0.63 for S-T-STR, D-T-STR, and T-T-STR, respectively. The ratio of maximum to minimum LLE values ranges from 2.22-3.98, 2.49-4.17, and 2.36-3.05 for S-T-STR, D-T-STR, and T-T-STR, respectively, with average ratios of 2.90, 3.16, and 2.72. Compared to S-T-STR and D-T-STR, the fluctuations of LLE in T-T-STR at different rotational speeds tend to be more stable, indicating a more consistent overall chaotic behavior of the flow field. The differences in LLE values between different axial positions for S-T-STR, D-T-STR, and T-T-STR at different rotational speeds range from 9.61% to 21.34%, 7.64% to 33.60%, and 3.97% to 10.61%, respectively.
Figure 14 shows the variation of $MSE$ with rotational speed for S-T-STR, D-T-STR, and T-T-STR at different axial positions. S-T-STR and D-T-STR exhibit varying degrees of fluctuation in $MSE$ with changing rotational speed. Specifically, $MSE$ increases significantly with increasing rotational speed in S-T-STR, while it slightly decreases with increasing rotational speed in D-T-STR. However, T-T-STR does not show significant fluctuations in $MSE$ with respect to rotational speed. This indicates that increasing the rotational speed has a positive effect on enhancing energy transfer within the S-T-STR flow field and overcoming the hindrance caused by interface effects on energy and material transfer. In contrast, for D-T-STR, increasing the rotational speed intensifies the interaction between impellers, thereby impeding energy transfer within the flow field. There are numerical differences in MSE values among different axial positions for all three configurations, reflecting spatial instability in energy transfer. The average $MSE$ values for the same axial positions but different rotational speeds are approximately 0.99-1.11, 1.19-1.30, and 1.20-1.46 for S-T-STR, D-T-STR, and T-T-STR, respectively. The ratio of maximum to minimum $MSE$ values ranges from 1.42-3.64, 1.44-1.71, and 1.09-1.40 for S-T-STR, D-T-STR, and T-T-STR, respectively, with average ratios of 2.72, 1.56, and 1.24. Compared to S-T-STR and D-T-STR, the fluctuations in MSE for T-T-STR tend to be more stable at different rotational speeds, indicating a steady and
efficient energy transfer within the flow field. The differences in MSE values between different axial positions for S-T-STR, D-T-STR, and T-T-STR at different rotational speeds range from 8.41% to 12.29%, 8.06% to 10.03%, and 15.84% to 20.98%, respectively.

Figure 15 illustrates the variation of $K_2$ with rotational speed for S-T-STR, D-T-STR, and T-T-STR at different axial positions. The three configurations exhibit varying degrees of fluctuation in $K_2$ with respect to rotational speed, with S-T-STR showing the most pronounced fluctuations, followed by D-T-STR, while T-T-STR does not display significant fluctuations with changing rotational speed. For the same axial positions but different rotational speeds, the average $K_2$ values are approximately 5.6168-5.6195, 5.6159-5.6181, and 5.6159-5.6167 for S-T-STR, D-T-STR, and T-T-STR, respectively. The differences between maximum and minimum $K_2$ values range from 0.22% to 0.37%, 0.11% to 0.22%, and 0.03% to 0.05% for S-T-STR, D-T-STR, and T-T-STR, respectively, with average differences of 0.28%, 0.14%, and 0.03%. Compared to S-T-STR and D-T-STR, the fluctuations in $K_2$ for T-T-STR tend to be more stable at different rotational speeds, indicating a smoother transition to a chaotic state within the overall flow field. The differences in $K_2$ values between different axial positions for S-T-STR, D-T-STR, and T-T-STR at different rotational speeds range from 0.0990% to 0.4706%, 0.0080% to 0.0409%, and 0.0015% to 0.0086%, respectively.
3.5 Comprehensive analysis

As shown in Figure 16, the interaction between the fluid and the impeller induces periodic torque fluctuations in the motor signal. This periodic energy input is reflected in the flow field, which exhibits chaotic characteristics of time instability overall but demonstrates periodic characteristics when the flow reaches a stable state, as observed in the Hilbert spectrum. Additionally, the spatial distribution of the impellers amplifies the differentiation in the flow field due to their interaction with the fluid, resulting in distinct flow field structures. Therefore, the overall chaotic performance and local spatial chaotic instability of the flow field are affected.

In the S-T-STR, D-T-STR, and T-T-STR configurations, interfaces between adjacent stirring blades, strong vortex regions at the center of the flow field, and annular vortex regions around the three impellers are formed, respectively. This leads to a more uniform flow field in T-T-
STR compared to the other configurations, as evidenced by the fractal dimension, \( LLE, MSE, \) and \( K_2 \) showing minimal differences along the axial positions. Furthermore, T-T-STR exhibits insensitivity to fluid viscosity due to the unique characteristics of its flow field. On the other hand, the presence of a strong vortex region in the center of the flow field in D-T-STR results in lower overall chaos and slower energy transfer compared to S-T-STR.

4 CONCLUSIONS

In this study, we conducted experiments in three different configurations of stirred reactors using glycerol-water solutions with varying viscosities. To overcome limitations for low-viscosity fluids, a novel approach combining flow field visualization with image processing was proposed to extract fractal dimensions and compare macroscopic parameters such as \( P_V, LLE, MSE, \) and \( K_2 \) to evaluate the chaotic behavior and energy transfer in the flow field. Furthermore, we utilized attractor images and the Hilbert-Huang transform to extract macroscopic and local temporal instability features from nonlinear physical quantities.

Comprehensive analysis indicated the existence of chaotic characteristics and inconsistent energy transfer in different axial positions of the S-T-STR, D-T-STR, and T-T-STR flow fields. T-T-STR displayed a weaker spatial instability compared to other configurations, consistent with the conclusions drawn from the flow field fractal dimensions. These findings further validate the reliability of our proposed flow field visualization technique for low-viscosity fluids. Furthermore, the relevant results also revealed time instability and chaotic characteristics in all three configurations, transitioning to periodic features as the flow field reached stability. Overall, T-T-STR exhibited weaker time instability, denser periodic cycles, and stronger evolution of the flow field structure and energy transfer rate. These distinctive
features are determined by the variations in flow field structures. Based on our flow field
visualization and experimental data analysis, we infer that the S-T-STR, D-T-STR, and T-T-
STR flow fields comprise adjacent impeller interfaces, strong vortex regions in the flow field
center, and annular vortex regions surrounding the three impellers. These findings can be
confirmed by additional flow field simulations.

AUTHOR CONTRIBUTIONS

Tong Meng: Experiment; software and procedure; data processing; writing–original draft.
Songsong Wang: Experiment. Qian Zhang: Writing–review and editing. Jie Yang: Software
Project administration. Changyuan Tao: Writing–review and editing. Zuohua Liu:
Supervision; writing–review and editing.

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DATA AVAILABILITY STATEMENT AND REPRODUCIBILITY STATEMENT

Due to the extensiveness of the experimental data in this study, all data has been processed and
is provided in the form of datasets available as spreadsheets.
The numerical data from Figures 4, 5, 6, 10, 11, 12, 13, 14, 15 and Supplementary Information Figure 2 are available as a data.zip file in the Supplementary Material. The Figures 7, 8, and 9 are generated by a custom-written program for direct data processing. Due to confidentiality reasons, we cannot provide the program itself. However, the original experimental data is available in a data.zip file provided in the Supplementary Material.

The other original data that support the findings of this study are available on request from the corresponding author.

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