Macro-instability: a chaotic flow component in stirred tanks

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Chaotic features of the macro-instability (MI) of flow patterns in stirred tanks are studied in this paper. Datasets obtained by measuring the axial component of the fluid velocity and the tangential force affecting the baffles are used. Two geometrically identical, flat-bottomed cylindrical mixing tanks (diameter of 0.3 m) stirred with either pitched blade turbine impellers or Rushton turbine impeller are used in the experiments, and water and aqueous glycerol solutions are used as the working liquids. First, the presence of the MI component in the data is examined by spectral analysis. Then, the MI components are identified in the data using the proper orthogonal decomposition (POD) technique. The attractors of the macro-instability are reconstructed using either the POD eigenmodes or a method of delays and finally the attractor invariants are evaluated. The dependence of the correlation dimension and maximum Lyapunov exponent on the vessel operational conditions is determined together with their distribution within the tank. No significant spatial variability of the correlation dimension value is observed. Its value is strongly influenced by impeller speed and by the vessel–impeller geometry. More profound spatial distribution is displayed by the maximum Lyapunov exponent taking distinctly positive values. These two invariants, therefore, can be used to locate distinctive regions with qualitatively different MI dynamics within the stirred tank.

Keywords: stirred tank; macro-instability; chaos; attractor; correlation dimension; Lyapunov exponent

1. Introduction

Stirred vessels are common in most of the process industries where they are used for a variety of purposes (to blend miscible liquids, to disperse a gas into a liquid, to suspend solid particles, to enhance mass and heat transfer, etc.) Flow phenomena encountered in stirred tanks are therefore a subject of numerous theoretical and experimental studies.

The flow of a batch in a stirred tank is highly complex and comprises components vastly differing in their temporal and spatial scales. The spatial scales of particular flows in the vessel extend from sizes comparable to vessel
dimensions (e.g. circulation loops) to microscopic turbulent eddies dissipating the kinetic energy of the liquid. Liquid flow in a stirred vessel hence can be viewed as a pseudo-stationary, spatially distributed dynamical system with high dimension of its phase space. The global liquid flow consists of a high number of hierarchically ordered unsteady and pseudo-periodic flows—circulation loops, vortices and eddies. The lifetimes of these flows span several orders of magnitude.

A decade ago, a particular pseudo-periodic macro-scale flow was identified in stirred vessels, manifesting itself on a spatial scale comparable to the size of the vessel and occurring with characteristic frequencies significantly lower than the impeller frequency. This flow was named the macro-instability (MI) of the flow pattern. The existence of MI has been confirmed by various experimental methods (e.g. Kresta & Wood 1993; Brüha et al. 1996; Montes et al. 1997; Bittorf & Kresta 2000; Nikiforaki et al. 2003). The presence of the MI in the flow pattern is displayed by distinct peaks in the low-frequency part of the power spectrum of a signal measured in the tank.

The macro-instability of the flow pattern has a strong impact on all mixing processes linked to fluid motions. It, however, also exerts strong forces acting on solid surfaces immersed in a stirred liquid (baffles, cooling and heating coils, etc.; e.g. Kraténa et al. 2001; Hasal et al. 2004). Detailed knowledge of the formation and time evolution of MI and a quantitative description of it are therefore of great practical importance. Until now most of the experimental efforts have been focused on the frequency of occurrence of the macro-instability and the MI kinetic energy. Our earlier analyses (cf. Hasal & Foršt 2000; Hasal et al. 2000), however, have also pointed to the chaotic nature of the macro-instability fluid flow component. It was suggested that MI is probably generated by a low-dimensional dynamical system with deterministically chaotic dynamics.

Methods of nonlinear analysis are applied to the analysis of experimental data from stirred tanks relatively scarcely, probably due to the high spatio-temporal complexity of the fluid flow (stochastic turbulent flows) encountered in stirred vessels. Ottino et al. (1992) described the mixing processes in fluids in terms of a general theory of chaotic processes and suggested links between them and their engineering counterparts. Letellier et al. (1997) analysed the velocity field in a standard mixing vessel with a Rushton turbine impeller. They introduced a mathematical procedure, based on an application of the Hilbert transform, enabling the separation of the deterministic component from the stochastic component of measured data. An attempt to analyse data from mixing tanks by nonlinear methods was performed also by Matsuda et al. (2003).

2. Experimental

(a) Stirred tanks and experimental conditions

Two geometrically identical, cylindrical flat-bottomed mixing tanks with four radial baffles (figure 1) were used in the experiments. Two series of experiments have been performed: the axial component of liquid velocity was measured in a region close to the impeller in the first series and the tangential component of the force affecting the baffles was studied in the second series.

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Velocity measurement

The axial component of liquid velocity in the stirrer region was measured by laser Doppler velocimetry (LDV) using a pitched blade impeller pumping liquid towards the vessel bottom and located at the off-bottom clearance of $C = 0.35T$. Water and two aqueous glycerol solutions were used as working liquids. Three values of the impeller Reynolds number, $Re_M = \frac{\varrho ND^2}{\eta}$, have been attained: 750; 1200; and 75 000. The flow regime in the tank therefore varied from strictly laminar to highly turbulent. In equation (2.1), $D$ is the impeller diameter (m); $N$ is impeller speed of rotation (s$^{-1}$); and $\varrho$ and $\eta$ are liquid density and viscosity (kg m$^{-3}$, Pa s), respectively. Details on the experiments can be found in Montes et al. (1997), Hasal & Fort (2000) and Hasal et al. (2000).

(c) Force measurement

The tangential component of the force affecting the baffle was measured by a mechanical device described in detail by Kraténa et al. (2001) and Hasal et al. (2004). The force was measured at several vertical distances, $H_T/H$, from the tank bottom (cf. figure 1). At each target position the value of $Re_M$ was varied over the entire accessible range. The study was performed using either pitched blade impellers (with six or four blades, pitch angle 45°, pumping downwards) at two off-bottom clearances, $C/H = 0.2$ and 0.35, or the Rushton turbine impeller at off-bottom clearances $C/H = 0.35$ and 0.5. Water and cold and hot aqueous glycerol solutions were used as working liquids. A range of $Re_M$ values from 16 000 to 83 300 was achieved with the pitched blade turbines (PBTs). The attainable range of $Re_M$ values for the Rushton turbine (RT) was 6000–63 000.
3. Experimental data analysis

Raw measured signals in all experiments were converted to digital form (possibly using a calibration equation) and numerically resampled with the sampling period $T_S = 20$ ms. Resulting time series were used for subsequent analysis. The procedures used for data analysis are only briefly mentioned here as the details can be found elsewhere (e.g. Broomhead & King 1986; Aubry et al. 1991; Hasal & Fort 2000; Hasal et al. 2000, 2004). Recently, we have established a technique enabling one to detect the macro-instability in measured time series, evaluate its relative magnitude and reconstruct its temporal evolution (Hasal et al. 2000, 2004).

First, the presence of the macro-instability in the data is detected and its frequency determined using power spectra of the experimental time series. The spectra are computed using an FFT-based (fast fourier transform) algorithm (Press et al. 1992). The procedure used for the extraction of the MI-related component of the signal consists of an application of the proper orthogonal decomposition (POD)—e.g. Broomhead & King (1986) or Aubry et al. (1991)—combined with spectral analysis. The measured data in this procedure are decomposed into a set of eigenmodes and eigenvalues. The value of the $k$th eigenvalue expresses the relative contribution of the $k$th eigenmode to the total variance of the analysed signal. In the case of velocity data, the variance equals the kinetic energy of the flow fluctuations. By (properly) summing the eigenmodes, the time evolution of the macro-instability-related component of the analysed data can be reconstructed. The eigenmodes contributing to the macro-instability are chosen from the entire set using their power spectra: the eigenmodes with only a single peak in their power spectra, matching exactly the macro-instability frequency (already determined by spectral analysis of the original data), are used. This way of resolving the frequency components of the data has proved to be quite efficient when applied to diverse datasets (Hasal & Fort 2000; Hasal et al. 2000, 2004). The chaotic attractors of reconstructed (extracted) macro-instability-related components of the measured data were reconstructed using either the POD eigenmodes or the method of delays. The embedding dimension was determined using the false nearest neighbour analysis. The time delay value was determined from the position of the first minimum of the mutual information function. Values of the maximum Lyapunov exponent were determined using the TISEAN package procedures (Hegger et al. 1999). Correlation dimension values were obtained using the Sano and Sawada algorithm (see Sano & Sawada 1985).

4. Results and discussion

So far, we have concentrated in our analyses predominantly on the reconstruction of chaotic attractors of the macro-instability-related components of measured fluid flow data and on the evaluation of their chaotic invariants. We make use of the correlation dimension in order to characterize the temporal complexity of the MI flow component at a particular experimental point, and further we evaluated the maximum Lyapunov exponent value as it measures the speed of divergence of trajectories in the phase space and therefore it may indicate the speed (magnitude) of dispersive processes produced by the macro-instability at a given location within the tank.
Vertical profiles of correlation dimension and maximum Lyapunov exponent values for chaotic attractors of the MI component of the axial fluid velocity are shown in figure 2. The plots in the figure show values of both invariants averaged across horizontal levels of experimental points (cf. figure 1) in order to suppress the fluctuations of individual values. The correlation dimension value (figure 2a) shows only minor variability with vertical position in the vessel. The effect of the impeller rotational speed is much more pronounced. The dynamics of temporal evolution of the MI component of fluid velocity is, therefore, almost coherent over the entire experimental region. The vertical profiles of the maximum Lyapunov exponent value (figure 2b) show more variability. Not only is the effect of the impeller speed again obvious, but also remarkable differences between the $\lambda_{\text{max}}$ values in the above and below impeller regions are evident. This difference arises from the different structure and properties of fluid streams in these two regions (e.g. Kresta & Wood 1993). Figures 3 and 4 give more detailed views of the spatial distributions of the $\lambda_{\text{max}}$ and $d_c$ values over the grid of the experimental points.

The distributions of $\lambda_{\text{max}}$ values in figure 3 obviously reflect the principal influence of Reynolds number ($Re_M$) value on macro-instability dynamics: at very high intensity of mixing ($Re_M = 75\ 000$, figure 3c) the macro-instability flow component is destroyed (to a remarkable extent) by the stochastic turbulent flows. The extraction of the MI component of fluid velocity is less reliable at these points and so is the evaluation of chaotic invariants of its attractors (considerable scatter of points in figure 3c). The $\lambda_{\text{max}}$ distributions are more homogeneous at lower $Re_M$ values (cf. plots in figures 3a,b). Higher values of the maximum Lyapunov exponent in the above impeller region of measuring the points are evident. When the mixing tank is designed for a dispersive process, the inlet ports or pipes would be introduced just into this above impeller region.
The correlation dimension, \(d_c\), exhibits quite homogeneous radial distributions (figure 4), i.e. its values do not change much with the \(r/R\) value (except for the scattered distribution evaluated at \(Re_M = 75\,000\)). The dependence of the \(d_c\) values on vertical position, \(z/H\), is more pronounced. The above and below impeller regions clearly differ in \(d_c\) value. The structures of state spaces of dynamical systems generating the MI flows in below and above impeller regions are therefore different and reflect well-known different structures of liquid flows in these regions.

The vertical distributions of the maximum Lyapunov exponent of attractors of the MI component of the tangential force affecting the radial baffle are shown in figure 5 for three different impellers at the same off-bottom clearance, \(C/H = 0.35\), and at various impeller speeds. The plots in figure 5—despite a remarkable scatter of the points—reflect the basic structure of the fluid flows in mixing tanks equipped with the impeller types used in our experiments. The PBTs induce single-loop circulation patterns with only relatively small differences between the above and below impeller parts of the liquid batch in the tank. As a consequence, the plots in figure 5(a,b) exhibit relatively low vertical variability, the only exception being seen in the below impeller region with the 6PBT impeller.

Figure 3. Spatial distribution of maximum Lyapunov exponent, \(\lambda_{\text{max}}\), of chaotic attractors of macro-instability component of axial liquid velocity at three impeller Reynolds number values: (a) \(Re_M = 750\), (b) \(Re_M = 1200\) and (c) \(Re_M = 75\,000\). Different symbols are used only to distinguish horizontal levels of measuring points (cf. figure 1).

Figure 4. Spatial distribution of correlation dimension, \(d_c\), of chaotic attractors of macro-instability component of axial liquid velocity at three impeller Reynolds number values: (a) \(Re_M = 750\), (b) \(Re_M = 1200\) and (c) \(Re_M = 75\,000\). Different symbols are used only to distinguish horizontal levels of measuring points (cf. figure 1).

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where $l_{\text{max}}$ takes markedly lower values. Here, the impeller discharge stream (probably) destroys the macro-instability flow. In contrast, the Rushton turbine impeller induces a two-loop circulation pattern in the tank, where several vertically distributed regions can be identified differing in the direction of flow and in the turbulence intensity. The graphs in figure 5c roughly reflect this flow structure by the ‘wavy’ character of the plots. The effects of the impeller rotation frequency are also obvious—higher impeller speeds lower the $l_{\text{max}}$ values as the MI component of the flow becomes more disturbed by the turbulence.

The vertical distributions of the correlation dimension values of the attractors of the MI component of the axial force affecting the baffle—depicted in figure 6—demonstrate (in a similar way as the plots in figures 2a and 4) that the correlation dimension takes an almost constant value along the baffle at fixed impeller rotation frequency. This fact suggests that the liquid jet ascending along the leading edge of the baffle represents, in principle, a quite homogeneous macro-flow occupying a substantial part of the vessel height. It corresponds to the basic macro-instability feature mentioned in §1. Hence, any apparent variability in spatio-temporal complexity of the macro-instability trajectories within respective attractors (and therefore also of the $d_c$ value) can hardly be anticipated. The plots in figure 6 indicate that the impeller rotation frequency exhibits, in general, only marginal effects on the correlation dimension value. Slightly more pronounced effects of the impeller speed on the $d_c$ value can be seen only in figure 6a, i.e. for the six-bladed pitched blade impeller that produces more distinguished flow patterns above and below the impeller. This difference between the two flow regions is reflected in an apparent local decrease of the $d_c$ value at $z/H=0.35$, i.e. at the impeller location where strong radial turbulent

Figure 5. Vertical distribution of maximum Lyapunov exponent, $l_{\text{max}}$, of chaotic attractors of tangential force affecting the baffle. (a) Six-bladed pitched blade impeller, (b) four-bladed pitched blade impeller and (c) Rushton turbine impeller. Cold glycerol solution was used as working liquid in all cases, with impeller off-bottom clearance $C/H=0.35$. Impeller speed of rotation: open circles, 390 min$^{-1}$; filled circles, 430 min$^{-1}$; open diamonds, 470 min$^{-1}$; filled diamonds, 510 min$^{-1}$; open squares, 550 min$^{-1}$ for PBTs; and open circles, 200 min$^{-1}$; filled circles, 240 min$^{-1}$; open diamonds, 280 min$^{-1}$; filled diamonds, 320 min$^{-1}$; open squares, 360 min$^{-1}$ for RT impeller. The thick line denotes the vertical position of the impeller blade lower edge.

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Liquid flow appears. This flow, however, is fully suppressed in the case of the four-bladed pitched blade impeller (figure 6b). The structure of flows within the stirring tank is more complex in the case where the Rushton turbine impeller is used for stirring. This complexity is reflected in more pronounced variability of the $d_c$ value along the baffle as shown in figure 6c.

The values of the correlation dimension reported in figures 2, 4 and 6 indicate low dimensionality of the dynamical system generating the macro-instability-related component of the liquid flows in stirred tanks—the $d_c$ value varies between 2 and 3 (the embedding dimension for reconstruction of the attractors varied from 3 to 4). Low-dimensional approximations to the macro-instability flows can therefore be constructed using, for example, the Galerkin approximation.

The mixing properties of the liquid flow in stirred tanks due to the macro-instability of flow pattern can also be characterized by the Shannon entropy production rate evaluated for the attractors reconstructed from the macro-instability components detected at particular flow conditions. The vertical distributions of the entropy production rate of the macro-instability-related component of the force affecting the baffle are shown in figure 7. As the estimates of the Shannon entropy are more robust, the plots in figure 7 are fairly smooth and resemble, in principle, the structure of the plots in figure 6. Two regions of flow (below and above impeller regions) in the tank are clearly visible for the six-bladed pitched blade impeller (figure 7a). The entropy production rate is somewhat higher in the below impeller region where the liquid flow is more vigorous when compared with the above impeller region. The effects of the impeller rotation frequency on the entropy production rate are rather insignificant. The same is true for the four-bladed impeller (figure 7b), which

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exhibits almost uniform vertical profiles of the entropy production rate at all stirring speeds. Again, this fact can be assigned to more homogeneous flow and turbulence fields achieved with this impeller type. The Rushton turbine impeller produces higher turbulence levels in the tank and this fact is mirrored in increased entropy production rate (figure 7) compared with the pitched blade impellers. The impeller rotation speed again poses only marginal effects on entropy production rate and its vertical distribution is fairly homogeneous.

5. Conclusions

The analyses performed in this work clearly demonstrated the chaotic nature of the macro-instability of the flow pattern in mixing tanks used in the experiments. Despite the large scatter of their experimental values, spatial regions with distinct dynamical behaviours can be detected within the stirred tank using the maximum Lyapunov exponents and the correlation dimensions of chaotic attractors of the macro-instability component of measured data. The correlation dimension of the MI attractor is suitable for the analysis of transitions between different flow patterns in the tank and of different MI types resulting from these transitions. The maximum Lyapunov exponent (taking distinctly positive values) can be used for finding regions with different MI dynamics within the tank and thus for finding the proper location of inlet and outlet ports of continuous flow mixers, location of gas spargers or analytical probes, etc.

Support of this project by the Czech Science Foundation (grant no. 104/05/2500) and the Czech Ministry of Education (Grant: MSM 6046137306) is gratefully acknowledged.
References


