Visualization of hydrodynamic and physico-chemical processes in rotating and vibrating containers

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Abstract. Variable inertial fields are an efficient way to control the behaviour of hydrodynamic systems. Forces of inertia can be used, for example, to stabilize or destabilize systems with an interface or density gradient, to mix multiphase or non-isothermal fluids. The implementation of this approach means that liquids fill the periodically moving containers. In this paper, the situations are considered when the containers perform either rotation or translational vibrations. Methods for measuring the density and velocity fields of convective flows in reacting hydrodynamic systems are described. Interferometry is used to visualize the density distribution. Particle image velocimetry (PIV) is used to study the structure and velocity of the flows. Optical instruments are installed stationary in the laboratory system. For video recording, a camera shutter is synchronized with the motion of a container, and thus the images are captured in a fixed phase of oscillations or rotation. Constructions of the containers make it possible to illuminate the working volume through transparent walls at different angles or in different planes. They also provide a compensation for the centrifugal pressure and allow interference cells to be used in overload conditions. The successful application of the methods in experimental studies of chemo-hydrodynamic processes is demonstrated.

1. Introduction
The forces of inertia provide a possibility to act on hydrodynamic systems that fill closed containers, for instance, such systems that have inhomogeneous density distribution. Among others, these effects are encountered in thermal vibrational convection [1] and may be illustrated by recent experimental and theoretical studies on the action of rotation or vibration on non-isothermal fluids [2, 3].

Vibration can dramatically change the dynamics of mechanical systems [4], for instance, a simple mechanical pendulum acquires new, otherwise impossible, steady positions if its point of suspension vibrates with the sufficiently high intensity [5]. This phenomenon is known as dynamic stabilization. For instance, it allows to suppress the Rayleigh-Taylor instability on liquid surface/interface [6], partially stabilize contact zone between miscible liquids [7], prevent a sedimenting liquefied granular medium from falling in particles [8]. As it was discovered in theoretical [9, 10] and experimental [11] works, vertical vibrations modify the regime of thermal convection and can even suppress it. On the other hand, when the frequency is sufficiently low and the amplitude is high, vibrations excite parametric resonance. Examples of it in stratified fluids may be found in [12, 13].

Rotating fluids demonstrate non-trivial behavior due to the action of inertia: Coriolis and centrifugal forces [14]. An example of application of rotation in chemical systems is the Taylor-vortex reactor [15, 16]. Earlier
studies of non-isothermal rotating fluids demonstrate that the interaction between the gravity and the centrifugal field leads to the onset of thermal vibrational convection [17]. Otherwise, by setting the study in a Hele-Shaw (HS) cell it is possible to exclude the gravity impact. In experiments it is achieved by orienting the rotation axis of the HS cell vertically, as it is going to be shown in the present work. Recent theoretical works [18, 19] demonstrate the perspectives of this approach. In contrast to vibration which creates an inertial field inhomogeneous in time, the rotation creates a constant in time, but spatially inhomogeneous centrifugal field. Thus, the convective structures not only evolve in time, but, as they move in the container’s volume, they are also affected by forces that vary in space.

Among various heterogeneous systems, the reacting fluids stand out. They not only have inhomogeneous density distribution, which alone already gives possibility of efficiently acting on the system by inertial fields. But they are also non-stationary, because the reagents are burning out with time and the problem formulation often corresponds to the batch operational mode of a reactor. Thus, we deal with systems, in which the mass forces acting on fluids may vary, as well as the boundary conditions. For instance, during the frontal neutralization reaction the latter are continuously varying at the zone of reagents contact.

The classic A+B→C system is the most appropriate for studying of mentioned above mechanisms of instability. In such systems, chemically-induced changes in the reagents properties can lead to the formation of hydrodynamic instabilities. Depending on reactor geometry and miscibility of reagents, surface-tension [20], buoyancy [21] and viscosity-driven [22] instabilities are known. The often studied reaction among the A+B→C types is the second-order exothermic neutralization reaction of a strong acid with a strong base. Although the reaction has simple kinetics, it has a nonlinear spatio-temporal evolution that explained its actively studying in recent years [23, 24]. For these reasons, we also consider chemoconvective processes in reactive system with the frontal neutralization reaction.

In the present work, various experimental optical methods are considered that are applied for the study of the flows and the density distribution in containers performing periodic motion. As objects of the study, two-layered systems of reactive miscible liquids are chosen. In the considered experiments, Hele-Shaw (HS) cells of different configuration are used. The focus is made on the interaction between chemoconvective patterns and inertial fields.

2. Experimental methods

Methods considered in the present work allow studying the hydrodynamics of heterogeneous systems in non-inertial frames of reference that are related to the moving containers. This is achieved by means of synchronization and image capture in a single, constant phase of rotation or oscillations. The details of the experimental realization of this approach are discussed in this section.

2.1. Setup with vibrating cell

In experiments a rectangular HS cell is used, which has the following dimensions: width $L = 50.0\;\text{mm}$, height $H = 100\;\text{mm}$, thickness $d = 1.2\;\text{mm}$ (figure 1(a)). The front and rear walls of the cell are made of high-quality, finely polished semi-reflective glasses (1) that are arranged parallel to each other due to calibrated glass insertions (2) installed in the corners (figure 1(a)). This way, an interferometer cell is formed. The vertical side walls are made of ordinary glass and kept transparent allowing for the entrance of a laser light sheet and, thus, rendering the cell suitable for the visualization by tracers. The HS cell is designed to study the chemoconvection in a two-layer system of miscible liquids that are characterised by different densities and diffusion coefficients. In most experiments, the initial density distribution is steady, i.e., a less dense liquid is situated above a denser one: $\rho_1 < \rho_2$ (figure 1(a)). The initial density profile is almost step-like. To achieve this, the following technique of cell filling is applied. The working volume can be accessed by channels, through four openings ($A, B, C, D$) situated in the middle of each narrow facet (figure 1(a)). The inlets $A$ and $C$ are used to feed the working liquids from the top and bottom, respectively. The outlets $B$ and $D$ are used to withdraw the mixed fluid from the contact zone. The liquids are injected and evacuated synchronously by means of a syringe pump. This allows creating a very thin mixing layer between liquids in the initial moment of time.
Figure 1. Diagram of the Hele-Shaw cell (a), diagram of the interferometer (c), photograph of the experimental setup (c): 1 – HS cell, 2 – insertions, 3 – frame, 4 – platform, 5 – shaker, 6 – accelerometer, 7 – pump, 8 – valve, 9 – laser, 10 – camera, 11 – interferometer, 12 – laser, 13 – micro objective, 14 – mirror, 15 – collimating lens, 16 – camera.
The HS cell is enclosed in a metal frame (3) and installed vertically on a movable platform (4) of an electrodynamic shaker 5 (figure 1(c)). To produce the high-frequency vibration of the cell, a modal excitation system Bruel&Kjaer of type 3628 is used. It is installed on a stand-alone concrete bed, disjoint from the laboratory floor. The operation of the shaker is controlled by means of a device Vibration Research VR 8500 and a set of multiaxial accelerometers (6). In the frequency range \( f = (10–100) \, \text{Hz} \) the experimental setup allows to vibrate the cell, shown in figure 1(c), with the peak overloads up to 40g. To fill the cell, a syringe pump (7) is used. It provides simultaneous motion of four syringes, two of which inject the working liquid solutions through the inlets A and C, while the other two extract the fluid from the zone of mixing. By adjusting the pumping rate it is possible to remove the mixed fluids and completely renew the contact zone between the initial solutions of working liquids. After the filling is complete, four valves (8), installed on each channel, are closed, and the vibration is switched on. When the shaker reaches the preset operational parameters: frequency \( f \) and amplitude \( b \), this moment of time is registered as \( t_0 \) and serves as a reference point for the evolution of the chemical system.

2.1.1. Visualization techniques. Three different methods are applied to study the flow velocity fields, the density distribution and the concentration of reagents. For the first one, polyamide tracer particles of neutral buoyancy Rilsan D40 (with the density 1.03 gr/cm\(^3\) and the size 40 \( \mu \text{m} \)) are added to both or either of the working liquids. The cell volume is then illuminated through the side wall with a light sheet created with a laser (9) (figure 1(c)). For this a continuous DPSS laser KLM-532/h/1000 with adjustable emission power (up to 1000 mW), equipped with a Powell lens and emitting in green (532 nm) is used. The images of the tracers are captured with a camera (10) through the front wall, and Particle Image Velocimetry (PIV) is then applied to obtain the fields of convection velocity. The images are processed with PIVlab software [25] and if necessary are analyzed in long series using the batch processing technique developed in experimental work [26]. Each image pair is processed in four passes with the consecutively decreasing interrogation window size, its side being equal 64, 32, 24, 20 pixels in the case of the Hele-Shaw cell presented in figure 1(c). The thickness of the light sheet is such that it illuminates the fluid across the entire depth of the HS cell. Consequently, the obtained velocity fields represent the velocity averaged across the layer thickness \( d \).

To study the field of density distribution, a home-made Fizeau interferometer (11) is used (figure 1(b)). The principal diagram of it is presented in figure 1(b). The interferometer incorporates a continuous 5 mW HeNe laser Plasma GN-5P (12) emitting in red (630 nm), optics (microscope objective (13), semireflecting mirror (14), collimating lens (15) and a camera (16). The difference in optical path is created between the walls 1 of the HS cell that are semitransparent. The interferometer is installed on a separate support to prevent vibrations. The interferograms are captured through the rear wall of the HS cell. The cameras are of models IDS UI-3250ML-M-GL and JAI CM-080GE and can be interchanged between the interferometer and the PIV setup.

The spatial distribution of reagents is studied with a colorimetric method using the universal color indicator. The presence of the indicator allows distinguishing media with different pH values. The universal indicator is a combination of several indicators (thymol blue, methyl orange, bromothymol blue, phenolphthalein, tropaeolin OO, bromocresol green and bromocresol purple) therefore it displays color changes over a very wide pH value range, from 1 to 10. Colors from orange to red indicate an acidic solution, colors from light to dark blue indicate bases, and colors from yellow to green indicate that the solution is neutral. Because the dynamic range of pH values of the indicator is wide, it is possible to visualize the variation of concentration even in the case when several components are dissolved at once. In the present study the reaction is frontal, so the salt formation occurs in a thin layer between the initial reagents. This zone is the reaction front. Since this place has the highest salt concentration, it is colored yellow (a medium close to neutral). During the reaction, the salt is mixed with the acid and the base, which is accompanied by a change in pH value and, accordingly, a change in the color of the medium that we can see and study in real time. However, obtaining the concentration field for one of the solutes becomes difficult. For this purpose, it would be more
appropriate to use an indicator that works in a narrow pH range. For example, the alkaline indicator allows you to visualize the field of alkali concentration and on the basis of the Lambert-Baer law to obtain quantitative values of its concentration. In this study, however, we use indicator only for visualization purposes. The indicator is added to the reagent solutions before the start of an experiment. In order to obtain a visualization of the pH distribution, the cell is illuminated by white collimated LED backlight (Metaphase Technologies). The image of the spatial distribution of the reagents is registered by CCD camera. The mass concentration of the indicator solution in reagents does not exceed 5%, and its presence, according to the earlier study [27], does not affect the structure and velocity of the convection.

2.1.2. Image capture. Synchronization. In order to exclude from consideration the oscillatory motion the fluid performs together with the cell, the images are captured in a single, fixed phase of oscillations. This means that the camera capture frame rate $f_i$ equals the frequency of vibration (rotation) $f$ or, in the case of a high speed motion, $f_i = 0.5f$. In order to avoid the image blur, the exposure time is kept equal or shorter that 1 ms. As a result, in the recordings the cell is seen immobile, and the visualizing patterns (tracers, color indicators, isolines of refraction index) are seen to move in the non-inertial frame of reference. This is realized by means of an optron-based device (17) (figure 1(c)) installed immovably near the vibrating platform (4). It generates electric tension upon the pass of an obturator fixed on the moving platform and sends a TTL signal to the camera’s logical input. The camera captures images only by the synchronizing signals. In the case of the rotating cell, a constant phase of rotation is fixed in a similar way. It is noteworthy that during PIV processing the fields of time-average velocity are obtained. The optimal time step within one pair is selected based upon a series of test calculations and, in practice, it usually has the order of magnitude within the range from 1 to 10 oscillation periods.

2.2. Setup with rotating cell

Experimental study of fluid systems dynamics in the field of centrifugal acceleration has a number of distinctions when compared to the case of vibration field. The methods of visualization remain the same: these are tracer-based method for velocity fields, indicator-based method for the distribution of the acid-base balance and interferential method for the fields of refraction index. The main technical challenge consists in the necessity to combine the strict requirement to the quality of geometrical properties of the interference cell (planarity and parallel alignment of the reference and test mirrors) with the large pressure of centrifugal origin in the fluid emerging at the high level of overloads. To overcome this contradiction, a special construction of the cell is used that has the form of a thick disk (figure 2(a)).

![Figure 2](image_url)

**Figure 2.** Photograph of the rotating cell (a) and its diagram (b): 1 – glasses, 2 – insertions, 3 – ring, 4 – spring, 5 – case, 6 – openings, 7 – liquid.
In figure 2(b), the diagram of the axial cross-section of such cell is presented. Glasses (1), which have the shape of disks with diameter 8 centimeters and are covered with a semi-reflective coating form a cell of Fizeau interferometer. The mutual parallel alignment of the glasses is assured by four glass insertions (2) that are bound together with a framing ring (3) and squeezed between the glasses (1). The thickness of these insertions determines the thickness of the working cavity of the cell, and the thickness inaccuracy does not exceed $\lambda/4$, where $\lambda$ is the laser wavelength. The thickness of the framing ring is a little bit smaller than that of the insertions; it is designed to prevent the insertions from moving. The clamping of the glasses to each other is assured by a conical spring (4). The interference cell is placed inside a dismountable case 5, made of acrylic glass. In the cell of this construction two cavities are formed: one cavity is between the glasses (1) and the other beneath them. These cavities communicate with each other along the exterior edge of the glasses. The excessive pressure in this situation does not wedge the glasses (1), but only pushes the upper glass. The maximum pressure is achieved on the cell periphery, where the edge of the glass (1) abuts upon the

![Figure 3](image)

**Figure 3.** Velocity fields obtained by PIV at the moments $t-t_0 = 4.2$ s (a), 281 s (b), 616 s (c), 891 s (d) during an experiment with vibration ($f=40.0$ Hz, $b=3.0$ mm) and a reagents pair HNO$_3$-KOH with molar concentrations $C_a = 3.0$ mol/L, $C_b = 4.0$ mol/L. In pictures, the central part of the HS cell is shown, the width of the presented area is 50 mm.
case (5), while in the central part of the glass the centrifugal overloads are small. Accordingly, the deformation of the upper glass is also small. Such construction allows to work at centrifugal overloads up to 103g on the cell periphery and obtain interferograms of high quality. The side wall of the case (5) is polished, which makes it possible to introduce a laser light sheet in the cell and apply the methods PIV and PTV to study the velocity fields.

On the axes of both glasses (1), filling openings (6) of different diameters are bored. This allows putting a filler tube from the syringe pump in either a position of filling the cavity I, or II and injecting fluid directly in the rotating container. There is also a possibility to simultaneously feed liquids through both filler tubes. When the tube is in position I the liquid fills the working cavity I between the glasses (1), flowing from the center towards the periphery. In position II, the liquid comes in the working cavity from the lower cavity and the filling occurs from the periphery towards the center. In the diagram, a cell partially filled with a working liquid (7) is shown. The use of such bidirectional filling is very practical in problems where the dynamics of heterogeneous liquid systems is studied. The initial state in such systems has the form of two half-volumes filled with different solutions and separated by a thin mixing zone.

![Image](image.png)

**Figure 4.** An interferogram of the convection driven by the CDD instability at $t - t_0 = 1265$ s (a); other experimental parameters correspond to those of figure 3. An example of the visualization by universal indicator (b): convective cells may be seen above the thin reaction front. The width of the presented area is 50 mm.

3. Chemoconvective flows in vibrating container

Let us consider the process of neutralization reaction between a solution of nitric acid, HNO$_3$, with the initial molar concentration $C_a = 3.0$ mol/L and a solution of potassium hydroxide, KOH, with the concentration $C_b = 3.0$ mol/L. The reacting system is subject to the vertical vibration with frequency $f = 40.0$ Hz and amplitude $b = 3.0$ mm, which correspond to the peak dimensionless overload $\Gamma = b \cdot 4\pi^2f^2/g = 19.3$. According to works [28, 29], at the considered initial concentrations of reagents, in the chemical system the concentration-dependent diffusion (CDD) instability develops. It is
characterized by the formation of a narrow horizontal row of convective cells above the reaction front. The evolution of the CDD instability in the field of vibration is shown in figure 3. The presented time-average velocity fields are calculated by PIV from the images of the central part of the HS cell. Initially, after the reagents are brought in contact, due to the appearance of reaction product, the diffusive layer convection (DLC) develops in the upper layer and the double diffusion convection (DDC) manifests itself in the lower layer (figure 3(a)). The flow structure evolves with the reaction (figure 3b) and due to the transformation of the density profile the upper layer is split in two domains: the convective cells emerge immediately above the reaction front and DLC plumes remain in the upper part of the volume (figure 3(c)). These two domains are separated with a relatively narrow diffusion layer (figure 4(a)). With time the height of the convective cells increases, meanwhile they continuously transform (figure 3(d)). At the same time, the reaction front progressively moves down. Below the convective cells, another diffusion layer is formed, as well (figure 4(a)). Following the classification from [29], the described convection belongs to the diffusion controlled (DC) regime.

![Figure 5. Interferograms of chemoconvection of CDD type in the rotating HS cell. Concentrations of reagents are as follows: NaOH – 2 mol/L, HCl – 3.3 mol/L. The rotation frequency is 10 rps. Two first frames are taken immediately before and after the injection of the upper reagent into the reactor. The width of the presented area is 40 mm.](image)

4. Chemoconvective flows in rotating container
As the object of study, the reaction between the hydrochloric acid and the sodium hydroxide was chosen. This choice was based on the analysis of experimental results from the studies of this chemical system in the conditions of constant Earth gravity. The following set of varied parameters was considered: rotation frequency 10 rps, initial position of the layers contact boundary in the middle of the reactor \( r = 2.4 \text{ cm} \), overload level on the initial boundary 10g. In figure 2, a series of interferograms demonstrating the evolution of CDD patterns for the considered case is shown. The first result that attracts attention is that the emergence of convective cells occurs considerably earlier than in the absence of the overloads. In video recording, a slow azimuthal drift of all convective
structures as a whole in the counter-clockwise direction is seen. Apparently, this is related to small rotary vibrations due to weak deviations in the rotation rate of the reactor. These vibrations do not produce any noticeable effect on the evolution of the convective structures. The further growth of the convective cells is accompanied by the increase of the wave number of the most dangerous instability mode. It was also accompanied by the expansion of the diffusion zone below the cells. The width of this zone and the concentration gradient within it were also noticeably larger than in the absence of the overloads. The further dynamics of this system will be determined by slow processes in the diffusion zone below the convective cells and can persist for a very long time. However, one can say for sure that the deficiency of the upper reagent will lead to that the reaction, as well as the convection, will soon terminate.

5. Conclusion
In the present work, we have proposed the methods of experimental study of the flows in non-inertial systems of reference: vibrating and rotating containers. The key feature in this case is the synchronized capture of the images. This approach is efficient when studying the slowly going processes and the flows averaged over the oscillation period. The optical methods that have been applied within the frame of this approach are interferometry, colorimetry and PIV. They allow visualization of the flow structure and of the distribution of reagents or medium density. Due to this, convective and physico-chemical properties, such as flow velocity, density and concentration, can be measured in the non-inertial systems of reference. Their dynamics can be tracked in time, as well.

The proposed methods have been demonstrated in the study of the dynamics of two-layer systems of miscible reacting liquids. They are also appropriate for the study of the thermal vibrational convection, which appears in oscillating non-isothermal fluids, for the study of steady streaming generated by the oscillations of a liquid–liquid interface in rotating containers or by the oscillations of liquid-filled container’s boundaries, and similar problems.

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References
[1] Gershuni G Z and Lyubimov D V 1998 Thermal Vibrational Convection (New York, USA: Wiley)
[2] Vjatkin A, Siraev R and Kozlov V 2020 Microgravity Science and Technology 32 1133–45
[3] Kozlov V, Rysin K and Vjatkin A 2021 Fluids 6 108
[4] Blechman I I 2003 Vibrational Mechanics (Delhi, Allied Publ.)
[5] Kapitza P L 1951 Physics-Uspekhi 44 7–20 (in Russian)
[6] Wolf G H 1969 Z. Phys. B227 291–300
[7] Wolf G G H 2018 Physics of Fluids 30(2) 021701
[8] Kozlov V G, Ivanova A A and Evesque P 2006 Fluid Dynamics and Materials Processing 2(3) 203–10
[9] Gel’fgat A Y 1991 Fluid Dynamics 26 165–72
[10] Carbo R M, Smith R W M and Poese M E 2014 The Journal of the Acoustical Society of America 135 654–68
[11] Swaminathan A, Garrett S L, Poese M E and Smith R W M 2018 The Journal of the Acoustical Society of America 144 2334–43
[12] Benielli D and Sommeria J 1998 Journal of Fluid Mechanics 374 117–44
[13] Bratsun D A, Stepkina O S, Kostarev K G, Mizev A I and Mosheva E A 2016 Microgravity Science and Technology 28 575–85
[14] Greenspan H 1968 The Theory of Rotating Fluids (University Press, Cambridge)
[15] Szczeszykowski J G, Koval C A and Noble R D 1995 Chemical Engineering Science 50(20) 3163–73
[16] Ramezani M, Kong B, Gao X, Olsen M G and Vigil R D 2015 Chemical Engineering Journal 279 286–96
[17] Vyatkin A A, Ivanova A A and Kozlov V G 2016 Journal of Applied Mechanics and Technical Physics 57(2) 344–51
[18] Utochkin V Yu, Siraev R R, and Bratsun D A 2020 Comput. Contin. Mech. 13(2) 205–18
[19] Utochkin V, Siraev R and Bratsun D 2021 Journal of Physics: Conference Series 1809 012017
[20] Tian R and Rongy L 2016 J. Chem. Phys. 145 124701
[21] Rongy L, Trevelyan P M J and De Wit A 2008 Physical Review Letters 101(8) 084503
[22] Gérard T and De Wit A 2009 Physical Review E 79 016308
[23] Eckert K, Acker M and Shi Y 2004 Physics of Fluids 16(2) 385–99
[24] Zalts A, El Hasi C, Rubio D, Urena A, and D’Onofrio A 2008 Physical Review E 77(1) 015304
[25] Thielicke W and Stamhuis E J 2014 Journal of Open Research Software 2 e30
[26] Mosheva E and Kozlov N 2021 Experiments in Fluids 62 10
[27] Mosheva E A and Shmyrov A V 2017 IOP Conf. Series: Materials Science and Engineering 208 012029
[28] Aitova E V, Bratsun D A, Kostarev K G, Mizev A I and Mosheva E A 2016 Journal of Applied Mechanics and Technical Physics 57 1226–38
[29] Mizev A I, Mosheva E A and Bratsun D A 2021 Journal of Fluid Mechanics 916 A22