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# Towards new contact-less techniques for the control of inertial particles dispersed in a fluid

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## Abstract

To achieve success in small-scale manufacturing processes and lab on chip applications new techniques and control principles are necessary. A promising technology to meet these objectives can emerge from improved understanding of induced particle self-organization mechanisms. Controlling the state of aggregation and properties of structures formed by particles immersed in a fluid is a subject of crucial importance at several scales. Practical applications range from the synthesis of advanced alloys to the crystallization of protein substances. Lack of adequate control in these processes is a fundamental methodological and theoretical shortcoming that limits applicability. Here we report on our recent discoveries about the possibility to use thermogravitational, thermocapillary or thermovibrational flows to force dispersed particles, regardless of their nature or type, to self-assemble and produce a variety of highly ordered, reproducible, high resolution structures. We show that the existence of these structures (also known as “attractors”) is due to the delicate interplay of different effects, which involve (but are not limited to) the inertial nature of the particles (i.e. their finite size and mass) and the existence of “waves” travelling in the considered fluid as a result of fluid-dynamic (Hopf) bifurcations or imposed (periodic) forcing. Some effort is also devoted to describe the topology and morphology of these attractors, which range from 1D helical curves to complex 3D surfaces depending on the specific conditions considered.

*Keywords: Inertial particles, thermocapillary, thermogravitational, thermovibrational flows, pattern formation.*

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## 1. Introduction

The study of the self-aggregation properties of particles dispersed in a fluid represents a rapidly growing field of research. This subject really stands at the intersection of many scientific branches, which make it a multi-domain field of investigations and a truly interdisciplinary science (combining many different apparently unrelated areas such as soft matter physics, engineering, biochemistry and even astrophysics [1]). From a practical standpoint, however, a general ability to predict precisely and scale-up rationally processes with particles dispersed in a fluid is still missing. Indeed, most of existing contactless approaches for the manipulation of dispersed particles are still based on the use of externally imposed magnetic or electric fields (for dilute fluid-particle systems) or on the ability of particles to stick together due to intrinsic inter-particle attractive forces, which make them very specific to the considered applications.

A possible alternative is based on the use of a ‘carrier flow’ to drive particles to specific regions of space or to promote networking events and self-organisation.

The ability of turbulence to support particle clustering or accumulation has been described over many years [2]. However, it has only recently been discovered that very common (non-turbulent, i.e. simply “laminar”) flows can produce very interesting and potentially useful phenomena in terms of patterning behaviour and particle structures [3-5]. While particle aggregates formed in turbulent flows have, in general, an irregular appearance (forming fractal-like structures [1]), these new classes of items display a variety of very regular shapes (for which in this study we coin the new denomination of “Highly Ordered Particle Structures” – HOPS).

The main overarching principle governing such phenomena is that, because of “inertial effects”, a set of particles when transported by a fluid can behave as a “compressible medium”, i.e. the spacing among particles can change significantly, even if the surrounding fluid and carrier flow are incompressible. This remarkable property can produce fluctuations of concentration of the dispersed matter and, hence, support mechanisms for particle accumulation and ordering. The subsets of space where particle clustering occurs are generally referred to as “attractors” (a terminology borrowed from the general field concerned with the study of non-linear systems). Even though the genesis of these phenomena can be associated to the inertia of particles, HOPS [5-11] do not conform to a simple definition or classification with respect to existing knowledge on particle accumulation phenomena. While inter-particle forces are not essential for their emergence, the presence of *waves travelling in the fluid* and their interaction with particles play a fundamental role in addition to inertia.

Travelling waves are ubiquitous phenomena. They arise everywhere in nature and a number of technological processes involve their formation at some stage. Here we show that HOPS can be formed by relatively simple (in terms of frequency spectrum) flows of natural origin, namely, thermogravitational or thermocapillary flows, which are initially steady, and become oscillatory in time (via a Hopf bifurcation) when the related control parameter (the applied temperature difference) exceeds a given threshold [6-8]. These flows can be still considered “laminar” and the associated mechanisms deterministic under a certain perspective.

We show that similar phenomena can also be promoted by the application of an external time-varying (periodic) ‘forcing’ to a non-isothermal system (leading to the so-called “thermovibrational convection” [9-11]).

## 2. Mathematical model

Direct numerical solution of the system model equations has proved to be a valuable means to test hypotheses about HOPS and obtain insights into the related dynamics. Such equations include the balance-equations for mass, momentum and energy, which in the framework of the Boussinesq approximation and in non-dimensional form simply read:

$$\nabla \cdot \underline{V} = 0 \quad (1)$$

$$\frac{\partial \underline{V}}{\partial t} = -\nabla p - \nabla \cdot [\underline{V}\underline{V}] + \text{Pr} \nabla^2 \underline{V} - \text{Pr} Ra T \underline{i}_g + \text{Pr} \gamma \beta T \sin(\Omega t) \underline{i}_\omega \quad (2)$$

$$\frac{\partial T}{\partial t} + \nabla \cdot [\underline{V}T] = \nabla^2 T \quad (3)$$

where  $\underline{i}_g$  and  $\underline{i}_\omega$  are the unit vectors along the direction of gravity and imposed vibrations, respectively and  $Ra = g\beta_T \Delta T L^3 / \nu \alpha$ ,  $\Omega = \omega L^2 / \alpha$ ,  $\gamma = b\omega^2 L^3 / \alpha^2$  (where, in turn,  $L$  is a characteristic length,  $\Delta T$  is the imposed temperature difference,  $\beta_T$  is the fluid thermal expansion coefficient,  $\nu$  is the kinematic viscosity,  $\alpha$  is the thermal diffusivity,  $g$  is the gravity acceleration,  $b$  is the amplitude of the considered vibrations and  $\omega$  their amplitude). An additional equation accounting for the balance of stresses at the free surface is required in the case of thermocapillary flow:

$$[(\nabla \underline{V}) + (\nabla \underline{V})^T] \cdot \hat{n} = -Ma (\underline{I} - \hat{n}\hat{n}) \cdot \nabla T \quad (4)$$

where  $\hat{n}$  is the unit vector perpendicular to the free surface, and  $Ma = \sigma_T \Delta T L / \mu \alpha$  is the well-known Marangoni number ( $\sigma_T$  and  $\mu$  being the derivative with respect to temperature of the surface tension and the fluid dynamic viscosity, respectively).

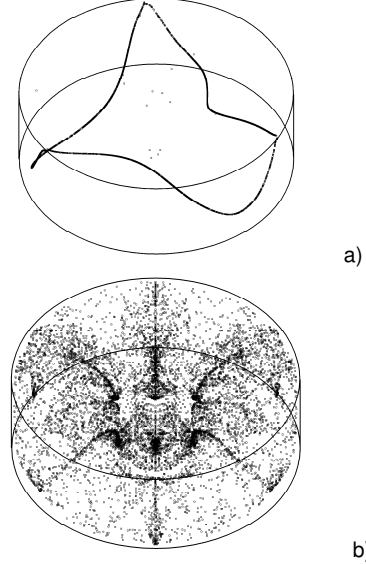
A separate Lagrangian tracking scheme is used for the particles, based on the so-called Maxey-Riley equation:

$$\frac{d\underline{V}_{partcl}}{dt} = \frac{1}{\xi + 1/2} \left[ -\frac{\text{Pr}}{\text{St}} (\underline{V}_{partcl} - \underline{V}) + \frac{3}{2} \frac{\partial \underline{V}}{\partial t} + \frac{3}{2} (\underline{V} \cdot \nabla) \underline{V} \right] + \frac{\xi - 1}{\xi + 1/2} \left[ Ga \text{Pr}^2 \underline{i}_g + \gamma \sin(\Omega t) \underline{i}_\omega \right] \quad (5)$$

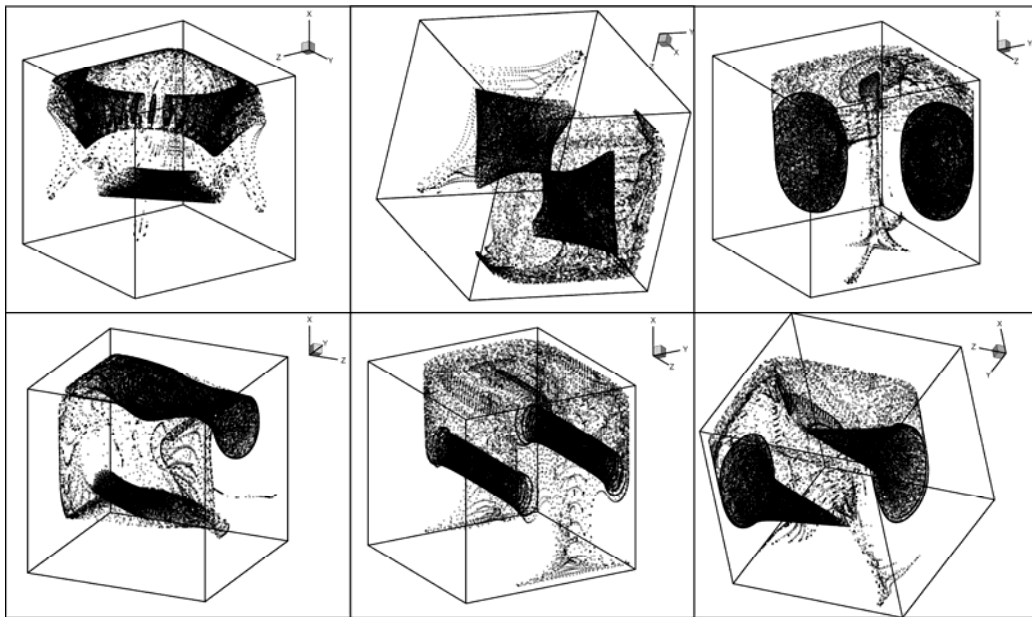
where  $\xi$  is the particle to fluid density ratio,  $Ga$  is the Galileo number ( $gL^3/\nu^2$ ) and  $St$  is the particle Stokes number  $St=2/9 (R/L)^2$ ,  $R$  being the particle radius. The Navier Stokes equations have been solved in the framework of a standard projection method. The emergence of HOPS has been considered in various geometries (cylinders and cubic containers).

## 3. Results

A kaleidoscope of particle structures are possible depending on the specific driving force considered. Beyond the specific situation or system considered, these structures are characterized by a recognizable level of self-organization (i.e. a precise morphology and/or topology in space and/or lines of evolution in time).



**Fig. 1.** Accumulation structures in a liquid bridge ( $Pr=8$ ,  $Ma=2 \times 10^4$ , height/diameter=0.34): a) 1D particle circuit formed by a travelling wave, b) Disconnected branches contained in distinct meridian planes emerging when the considered flow is a standing wave.



**Fig. 2.** Examples of particle structures (attractors) spontaneously formed by particles in a non-isothermal vibrated dilute liquid-particles system (encapsulated in a cubic container) for different relative direction of the imposed temperature gradient and direction of vibrations ( $Pr=8$ ,  $\Omega=10^3$ ,  $\gamma=1.5 \times 10^6$ ,  $\xi=1.85$ ,  $St=10^{-3}$ ).

Particle attractors range from one-dimensional circuits (with particles orderly aligned along a closed helical curve, 1D HOPS, see, e.g., Fig. 1a) to aesthetically appealing open or compact three-dimensional surfaces (2D or 3D HOPS, Figs. 2).

In the case of 1D HOPS, particles (initially uniformly spaced in the liquid), undergo segregation and form a seemingly rigid filament that moves as “a unit” (in general, in the form of a multi-blade windmill or a multi-sided star-shaped polygon, of morphology resembling the petals of a flower when projected in a specific plane of the fluid domain, Fig. 1a).

Most surprisingly, 3D HOPS resemble the so-called “quadrics” of projective geometry (such as cylinders, ellipsoids, saddle-horse paraboloids, and conical shapes) or amphora-like structures (some examples being shown in Figs. 2). Intermediate cases are also possible where the resulting pattern consists of disconnected 2D surfaces formed by particles, which apparently partition the considered fluid domain into different subregions (“like the pieces of a cake cut by a knife”, Fig. 1b).

Such results are being used for the preparation of dedicated space experiments (to be performed onboard the International Space Station: the so called JEREMI and T-PAOLA experiments). While the former (Japanese European Research Experiments on Marangoni Instabilities) will entirely be dedicated to the analysis of 1D and 2D HOPS emerging in liquid bridges, the latter (Thermovibrationally-driven Particle self-Assembly and Ordering mechanisms in Low gRAvity) will focus on the emergence of 3D HOPS in closed cavities and their sensitivity to frequency and amplitude of vibrations.

### 3. Conclusion

According to our numerical simulations, the subsets of space where particle are attracted evolve from perfect one-dimensional (1D) curves with particles orderly aligned along a closed helical wire or multi-blade circuits in the case of a single travelling wave, or planar (2D)

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areas for the case of two waves counter-propagating with equal amplitude, to three-dimensional (3D) geometric objects resembling the typical ‘quadrics’ of projective geometry, when single-wave or multi-wave flow patterns are replaced by vibration-induced oscillatory flow.

Future activities shall be devoted to test unifying hypotheses and interpretations for HOPS and to the practical exploitation of such a new knowledge.

The reason for our interest in such specific dynamics resides in the fact that, like magnetic fields, waves (produced naturally by the considered thermal flow or induced artificially through the application of vibrations) allow *contactless control of the dispersed phase*, but, unlike magnetic fields, they may be regarded as a new technique that can be used *more universally*, because its application is not limited to electrically conductive melts and/or particles.

The generality of such idea is made even more evident by the fact that, as no inter-particle forces are required to drive the ordering or accumulation process, it is also applicable to dilute systems and, in general, to circumstances where the particles do not display any intrinsic ability to stick together (there is no need for attractive electrostatic forces such as those that characterise macromolecular or colloidal substances).

Moreover, the fluid flow needs not to be in turbulent conditions, which implies the remarkable theoretical possibility to control particle dynamics *precisely* (via well-defined *laminar flows*, whose properties can be defined “a priori” on the basis of the problem geometry and “characteristic numbers”).

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