

EUROPEAN FEDERATION OF CHEMICAL ENGINEERING WORKING PARTY ON MIXING

Development, State of the Art and Future of the Scientific Field

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At present (July 2004), the Working Party on Mixing of the EFCE (established in 1978) consists of 30 delegates (members and standing guests) from European states who have contributed to this review and eight overseas delegates. The delegates of this Working Party are principally holders of academic positions, and consequently this overview is essentially devoted to research. It should be noted that, due to the small number of industrial delegates involved, information on the mixing trends from both a manufacturer's point of view and that of an industrial user (mainly from research centres) is rare. Every three years, the European Conference on Mixing is organized where academics and industrial partners involved in mixing research and development are accustomed to meet and exchange information and expertise, as well as to discuss open problems. The next conference, which will be held in Italy in 2006 (probably in June), will be the twelfth of the conference series. In Europe, at least 30 academic research laboratories are involved in the field of mixing, which indicates that mixing is a major topic amongst the various scientific areas of chemical engineering in Europe.

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STATE OF THE ART

Amongst the various unit operations in the domain of chemical engineering, some of the greatest progress in the last 20 years has probably been achieved in the field of mixing. Since this time there has been a significant step forward in the scientific knowledge of mixing, enabling a better understanding of the physical phenomena. A major consequence of this is the improvement of industrial processes and their efficiency, being marked particularly by increased process reliability, more reliable equipment scale-up and reduced product loss and wastes, thus contributing to overall economical progress.

This progress is due firstly to the outstanding development of the experimental methods available. With the advances in optical and laser devices, measurement techniques have become more and more accurate, and thus better adapted to the needs of mixing. The early techniques,

such as hot film anemometry, have almost completely disappeared. Nowadays, techniques employing lasers, such as laser Doppler anemometry (LDA), particle image velocimetry (PIV), planar laser-induced fluorescence (PLIF) and tomography are readily used in the research laboratories. These are extremely efficient techniques, enabling detailed information on hydrodynamics and mixing phenomena to be obtained. Nevertheless, the implementation of such experimental methods is not always so easy: firstly, such equipment is very expensive and involves a major financial investment; secondly, these techniques require a minimum level of expertise for set-up, fine tuning and application to mixing experiments.

The progress in the field of mixing is also strongly linked to the increased capability of computers and the subsequent development of commercial computational fluid dynamics (CFD) codes, such as ANSYS-CFX, Fluent and STAR-CD, which are the main codes used in industrial or scientific mixing. Until about 10 or 15 years ago, researchers themselves had to write their own simulation computational programs in order to simulate the hydrodynamics in a stirred vessel. Such programs were highly simplified, allowing the Navier–Stokes equations to be solved with a large number of assumptions and for very basic problems only. These calculations could take days and even months to

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reach convergence and a satisfactory result. Today, the hydrodynamics in a single phase stirred tank can be resolved within only a few hours, and in very close agreement with experimental results. Over recent years, the development of commercial codes has evolved rapidly, and the implementation and continual improvement of specific models (rheological, multiphase, chemical reaction, etc.) now enables the modelling of complex systems.

With the availability of these advanced experimental and numerical techniques, it is logical that studies devoted to mixing processes are no longer limited to single-phase systems and have turned towards more complex mixing problems. Elaborate research is now carried out in all areas of mixing: solid suspension, gas–liquid and liquid–liquid dispersion, viscous and rheologically complex fluids, chemical reactions etc. The progress made in these areas during recent years is very important and has evoked great interest from the process industries. Of course, the more fundamental, although vital, research on hydrodynamics in single-phase systems continues to be developed. The advances in both experimental and computational methods enable a better understanding of the mixing mechanisms and phenomena, especially with respect to turbulence, macro-instabilities and hydro-dynamic bifurcations.

SCIENTIFIC DEVELOPMENT

Single Phase Systems: Hydrodynamics and Turbulence

Owing to the strong progress in experimental and numerical techniques, single-phase mixing studies are still current research topics in most of the European laboratories involved in the field. These modern approaches are employed in addition to the more traditional studies, such as determination of power consumption, mixing time or RTD for continuous systems.

The development of the LDA and PIV techniques has led to huge scientific advances in the local characterization of hydrodynamics and turbulence. However, there are restrictions on the use of such techniques: they can only be performed on a small scale, and they require transparent vessels and optically clear liquids. The effects of system geometry and process conditions on the hydrodynamics in single and multiple impeller stirred tanks, as well as static mixers, have been investigated on a local scale, enabling the thorough characterization and performance evaluation of the various mixing systems. Other techniques, such as liquid crystal thermography, have been used for the precise determination of mixing times. PIV measurements have further enabled the identification and characterization of macro-instabilities in the flow which have also been detected using LDA and time series analysis: PIV seems to open another promising area. Even with these sophisticated measurement methods one quantity, the turbulence dissipation rate, remains elusive and yet many physical processes depend on knowing its local value. Some recent work has explored new methods for obtaining this quantity from instantaneous PIV velocity fields, but it is far from being a solved problem.

There has also been important progress in the area of modelling and CFD simulations of stirred tanks. The development of three-dimensional geometry modellers

means that complex impeller geometries and tanks can now be depicted realistically. The need for experimental data as input for the simulations (impeller boundary approach) is now obsolete. Alternative methods have since been developed, which also involve the resolution of the Reynolds averaged Navier–Stokes (RANS) equations. These include the sliding mesh and multiple reference frame approaches, which allow time-dependent and stationary modelling of stirred tank flow, respectively. Such methods provide flow fields which are in close agreement with the experimental data if sufficient mesh points are used, although there are some limitations with respect to the prediction of the turbulence quantities.

More recently, research groups have taken much interest in large eddy simulations (LES) for stirred reactors, enabling the unstable nature of the flow in stirred tanks to be modelled and better representation of the turbulence structure to be obtained. The limitation here is the calculation times, which are very long. It is also interesting to point out that the Lattice–Boltzmann technique appears to be a very promising method for both single- and two-phase flow simulations.

Solid–Liquid Systems

Solid–liquid suspension remains one of the most studied areas and almost all of the research laboratories in Europe which are active in the domain of mixing have an interest in this field. New experimental approaches allow the quantification of the liquid and solid velocities, as well as the turbulence level and its influence on the particle terminal velocity by the way of PIV, in addition to indirect methods. The degree of particle suspension in the vessel has been assessed using specific techniques based on pressure measurements. Studies on dense suspensions have also been carried out, whereby detailed measurements are now possible via X-ray tomography and image analysis. The effect of hydrodynamics on the particle size and morphology in precipitation reactions and flocculation has also been thoroughly investigated.

The vast amount of experimental data has enabled the development of phenomenological models and correlations for the description of the particulate phase distribution. Concerning CFD simulations, various models have been implemented in the commercial codes, such as simplified population balance models for precipitation and crystallization, Eulerian–Lagrangian and Eulerian–Eulerian models for the prediction of the particle distribution within the vessel, giving very close agreement with the experimental results. Nevertheless, in some cases, CFD simulations are not yet fully suitable for prediction of the correct behaviour and it is recommended to our colleagues involved in mixing studies that they be well aware of the limitations of the CFD codes.

Gas–Liquid Mixing

Gas–liquid mixing is another area which is well studied amongst the active research groups in Europe. This is due to both the progress in technology, enabling more detailed studies, as well as the vast field of application of gas–liquid reactors.

Traditional investigations on gas–liquid mixing systems, such as the determination of power consumption, gas holdup, flooding–loading transitions and mass transfer coefficients are still part of most experimental studies, since they enable the mixing system to be characterized in a global manner, which is now well understood. The influence of system properties (e.g. coalescing and non-coalescing) on the quality of gas dispersion, holdup and mass transfer coefficients has been analysed. The recent advances in the experimental technology, however, now enable the gas–liquid dispersions to be investigated in a more local manner, allowing precise information on both the dispersed and continuous phases to be obtained. LDA and PIV have been used successfully for the characterization of the bulk liquid flow patterns and the performance of the impeller in the presence of gas bubbles, in both non-viscous and viscous flows. An extension of the PIV technique, whereby the measurements have been synchronized with the passage of the impeller blade, has also enabled the study of the effect of the gas phase on the trailing vortices and the formation of gas cavities behind the blades. Regarding the gas phase, a number of techniques have been developed for the determination of local holdup, bubble size and velocity—these include capillary suction probes, optical probes, shielded conductivity probes, phase Doppler anemometry (PDA), and various methods of digital imaging.

The availability of such experimental data has hence resulted in detailed phenomenological models and correlations, enabling the description of the gas behaviour. The development of various multi-phase models in CFD codes has also encouraged the simulation of gas–liquid dispersions. Eulerian–Lagrangian, Eulerian–Eulerian and volume of fluid (VOF) approaches have been employed to model gas distribution in the stirred tank, as well as single and multiple bubble behaviour.

Many of the studies in gas–liquid mixing are specifically applied to bioreactors and biochemical engineering problems. The improved knowledge of the gas behaviour near and at the impeller blades has thus allowed the development of new impeller designs which are better suited to these applications. In this area, further studies include the effect of aeration, power input, mixing intensity and mechanical stress on the cultivation of micro-organisms and their morphology.

Gas–Liquid–Solid Mixing

Gas–liquid–solid mixing is additionally complex due to the presence of three phases in the flow. Nevertheless, new experimental techniques offer the possibility to study such systems, and investigations have been successfully carried out in a couple of the European laboratories. Studies range from the characterization of gas–liquid–solid and gas–liquid–fibre suspensions coming from the paper manufacturing industry using LDA, to the determination of bubble sizes and flow fields using PIV, particle emission tomography (PET) and positron emission particle tracking (PEPT) in catalytic oxidation and hydrogenation reactors.

Liquid–Liquid Dispersion

Until recently, this area has essentially been treated by physico-chemists or formulation specialists. Over the last few years, however, mixing has been recognized as a key parameter in the fabrication of liquid–liquid dispersions and emulsions.

Investigations generally consist in studying the effects of turbulent flow and the liquid composition (interfacial tension, viscosity ratio, presence of additives, etc.) on drop breakage and coalescence, where emulsion polymerization and phase inversion appear as the main applications. This research field is limited by the absence of specific measurement techniques that allow the *in-situ* determination of drop diameters in concentrated media. As a consequence, researchers often develop their own methods: LDA and PIV have been used to study dispersions of 10 and 25% dispersed phase by matching the refractive indices of the two liquids. More recently, a new technique using a stereoscopic probe has been proposed for *in-situ*, real-time measurement of drop size characteristics and distribution, which has also resulted in the discovery of phenomena such as drops-in-drops.

Owing to the complexity of the phenomena involved in drop break-up and coalescence, the capabilities of the commercial CFD codes are still limited in the predictions of emulsion formation or liquid–liquid dispersion. Nevertheless, work has been focussed on the development of various population balance models and multi-fractal turbulence models, as well as Eulerian–Lagrangian simulations.

Viscous and Non-Newtonian Mixing

Today, the mixing of viscous fluids and non-Newtonian fluids is studied mostly for specific applications. These include gas–liquid dispersions in bioreactors, polymerization reactors and equipment, highly concentrated solid–liquid suspensions, concentrated emulsions and particular chemical reactions in laminar or transient flow, amongst others. Among the basic studies, topics like power consumption, homogenisation characteristics and cavern formation are to be mentioned.

Particularly novel techniques, such as PETP and PET, are used to investigate the mixing of non-Newtonian liquids with and without particles, and to determine the extent of fluid volume that is agitated and solids distribution in complex opaque fluids. The implementation of such techniques in the research laboratories is, however, still quite limited. Other experimental studies involve the effect of laminar mixing on the course of chemical reactions, the visualization of reaction zones and the determination of residence time distributions.

Various tools, as well as theoretical and CFD models, are also currently being developed in order to predict mixing and micromixing in viscous systems, and to study the flow instabilities, which occur when liquids with significantly different viscosities are mixed.

Chemical Reaction

It is well known that the hydrodynamics and, more particularly, micromixing have an effect on complex chemical reactions. Typically, investigations have been carried out to

control the product distribution or selectivity in order to obtain pure products, thus avoiding or limiting the separation or purification stages. Furthermore, novel feeding strategies have been found to improve selectivity in fast chemical reactions. The study of precipitation reactors, where the product is a solid, has been developed in some laboratories. The attempt to tune the particle size by defining the agitation conditions has received some attention.

The interaction between mixing and chemical reactions (fast reactions in turbulent media and competitive parallel reactions) has also been modelled using CFD. Much work has been devoted to the development of mechanistic micro-mixing models, which, along with probability density function tools, have been coupled with CFD, allowing the prediction of reactive turbulent mixing.

Micromixers and Microreactors

The recent development of microreaction technology for the process industries has launched a new area of study in the field of mixing, concerning in particular micromixers and microreactors. Owing to the extremely small size of microreactors, the flow is almost always laminar and therefore mixing is difficult, as it is limited by the molecular diffusion mechanism. Experimental and numerical investigations are, however, not so simple: the experimental equipment and methods must be adapted to the minute size of the mixing system and the physical phenomena; numerically, the micro-sized reactors introduce additional difficulties with respect to the characterization and numerical diffusion. This implies that care is necessary in the choice of the discretization scheme and on the mesh resolution.

The development of the micro-PIV technique (PIV coupled with a microscope), as well as other digital imaging techniques, now enables the visualization and evaluation of mixing in microreactors. Studies have been conducted to investigate liquid mixing in microchannels, as well as gas-liquid dispersions and complex fluid mixing in meso-scale reactors for high-throughput screening applications.

CFD has also been used as a tool for the design and characterization of micromixers. Studies include the design of micromixer geometry and its effect on the mixing quality, the development of methods for the quantification of mixing, and miscible liquid chemical reactions.

FUTURE TRENDS

This short survey on the state of art in mixing shows that the mixing research in Europe spreads over all application fields, from bioreactors to suspensions to emulsions to chemical reactions, etc. Many of the studies carried out have received special grants from either national or European research programmes and others are supported by industrial funding. This active support is proof that mixing in the process industries is still a real problem and that a better understanding of the mixing mechanisms can improve process efficiency.

For many years mixing research was concerned with making global characterizations of flows and process operations in agitated vessels. The improvements and innovations in measurement techniques that now allow

local data to be obtained have strongly contributed to a better identification and understanding of the phenomena involved. Nevertheless, there is a deficit in real application of much fundamental understanding of the micro-scale processes that are involved. Other, more specialized, techniques developed in the laboratories are necessary for a deeper understanding of more particular cases.

CFD is generally used by the mixing community as a tool for the quantification of the behaviour of a system and its design. In very specific problems, such as rheologically complex fluids, multiphase flows or problems with complicated boundary conditions, existing models will need to be improved or new physical models will need to be developed, leading to a new contribution in CFD. The rapid evolution of technology and computers will also allow LES to become more accessible for the study of stirred tank problems, enabling an even deeper understanding of the mixing phenomena, turbulence and particle-drop-bubble dispersion at small scales.

A lot of the future trends in mixing will, of course, go hand in hand with the areas detailed above, with the aim of better understanding the mixing mechanisms involved in order to improve or to optimize the process. In addition, investigations in new research areas will be established in order to respond to the demands of the continually evolving process industries—research on some of these new topics has already begun in various groups.

Towards a Product-Oriented Chemical Engineering

Process design is now a concept which has been assimilated by the process industries. The mechanism of mixing has an important role in the conception of the final product and, according to initial or operating conditions employed, the properties of the product may differ. To attain this objective, the need for a deeper understanding of mixing processes and complex modelling capabilities will be even stronger. The definition of the quality of a product will perhaps become a key parameter. In parallel, it can be expected that improvements in experimental techniques and the development of new models will be necessary in order to link the required properties of the final product to the experimental data. In this respect, the ability to deal with rheologically complex fluids and to turn towards new applications areas, such as emulsification, polymerization, biomedical engineering, food or bioprocessing, will become more and more important.

Towards Nano-materials

One of the most challenging aspects of studying and modelling mixing is the fact that length and time scales involved in real processes vary by several orders of magnitude. This observation is particularly important in nanotechnologies. In the production of functional nano-size materials with large scale equipment, the interaction between small (micro- and nano-) scales and large scale are crucial to the understanding of the overall process.

Towards Micro-technologies

The development of micro-technologies has evoked much interest from the industrial community, mainly due to the process safety and intensification that they provide. In order to correctly implement micro-technologies in the process industries, new methodologies of design, numbering-up and characterization must be developed. Owing to the small scale of microreactors, the flow and process conditions deviate from what is traditionally understood in classical mixing problems, which opens a new domain in chemical engineering.

Towards Systems with New Design

The role of the agitator and of its surroundings is essential to the success of the mixing operation. Investigations must be carried out in order to develop systems with new and innovative designs, non-conventional vessels or agitation configurations which are better adapted to specific applications.

This also includes the need to know how to get the best out of existing equipment to manufacture all manner of new products, a flexibility which is often required by speciality chemical companies.

In addition, special care must be taken to study the mechanical aspects of mixing, such as the stress analysis of the

rotating impeller and shaft, possible vibrations due to inherent fluid-dynamic instabilities, the loading of the vessel and its internals (radial baffles, draught tubes) and erosion/corrosion phenomena in the mixing equipment.

REFERENCES

An overview of the main trends in mixing can be found in the proceedings of the last main conferences:

- *5th International Symposium on Mixing in Industrial Processes (ISMIP5)*, Seville, Spain, June 2004—a selection of the papers will be published in *Chemical Engineering Science*;
- *11th European Conference on Mixing*, Bamberg, Germany, October 2003—preprints edited by VDI-GVC, selected papers published in *Chemical Engineering and Technology* [27(3), March 2004];
- *4th International Symposium on Mixing in Industrial Processes (ISMIP4)*, Toulouse, France, May 2001—selected papers published in *Chemical Engineering Research and Design* [79(A8), November 2001];
- *10th European Conference on Mixing*, Delft, The Netherlands, July 2000—proceedings edited by H.E.A. van den Akker and J.-J. Derksen (Elsevier).

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