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Micro-mixing measurement by chemical probe in homogeneous and isotropic turbulence

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HIGHLIGHTS

- Validation of a micro-mixing model under homogeneous and isotropic turbulence (HIT).
- HIT is obtained by a system of oscillating grids placed in a vessel.
- Chemical probe measurements are carried out by the iodide/iodate reaction system.
- IEM and EDD models are tested.
- EDD model provides the more accurate quantitative characterization of micro-mixing.

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ABSTRACT

The chemical probe is commonly used to evaluate the performance of chemical reactors. By a localized injection of chemical reagents, it is possible to measure the local micro-mixing, which is readily related to the selectivity of chemical reactions, since mixing at the molecular scale is the limiting factor for a wide range of chemical systems. The raw result of the chemical-probe method is a segregation index that allows comparison of different situations (various locations in the reactor, various Reynolds numbers, various geometries, etc.). Beyond the qualitative assessment provided by this segregation index, it is possible to obtain the intrinsic micro-mixing time by means of a micro-mixing model, describing the temporal evolution of a chemical reaction whose rate is governed by the micro-mixing. Here a key step is the choice of micro-mixing model. Several micro-mixing models available in the literature have been used in some specific cases without evaluating their appropriateness for the problem in hand. The main difficulty in this evaluation is that the real flows often do not fully satisfy the basic model assumptions, in particular the condition of homogeneous and isotropic turbulence (HIT).

The present work aims at assessing the validity of a micro-mixing model under “ideal” experimental conditions, i.e. HIT with no mean flow, to avoid the bias due to the flow gradients. The HIT is obtained here by a system of oscillating grids placed in a vessel. The chemical probe measurements carried out by the iodide/iodate reaction system are applied to the two most commonly used phenomenological models in the literature: the IEM (Interaction by Exchange with the Mean) and the EDD (Engulfment, Deformation and Diffusion) models. The benchmark for the micro-mixing models is based on comparison of the local turbulent kinetic energy (TKE) dissipation rate both drawn from the micro-mixing time by the theoretical model of Bałdyga and the reference direct experimental determination by laser Doppler velocimetry measurements. It is shown that the engulfment model EDD seems the more appropriate to analyze the chemical data and provide a quantitative characterization of micro-mixing.

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0. Introduction

Understanding of the mechanisms that underlie turbulent mixing and convective transfer is fundamental in many technological applications – chemical, pharmaceutical, food, and cosmetic, for

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example [1]. Indeed, the selectivity of the fast chemical reactions depends strongly on the way in which the reagents are mixed on a molecular scale [2]. This mechanism is called micro-mixing, the ultimate scale of mixing by the flow before molecular diffusion comes to action. The kinetic of micro-mixing is a key parameter for the selectivity of the chemical reactions: micro-mixing must be “faster” than the chemical reaction to let the intrinsic reaction rate be effective [3].

Three parallel mechanisms can be distinguished in turbulent-mode mixing: macro-mixing, meso-mixing, and micro-mixing [4]. Macro-mixing takes place at the mixer scale and represents the fluid dispersion by the mean velocity fields. A fluid in a mixer disperses under the effect of the mean flow and is finally distributed in the whole mixer volume. This stage of mixing is influenced by longitudinal or transverse swirls that contribute to convective mass transfer and macro-mixing is characterized by the residence time distribution (RTD). In laminar flow macro-mixing can be improved by geometries that induce a chaotic advection, which makes the flow irregular [5,6].

In the presence of velocity fluctuations with no mean value, meso-mixing is related to the “random-path” turbulent agitation of fluid particles, analogous to a Brownian macroscopic motion. Nevertheless, it is governed not by a Boltzman statistics, but by a mechanism of scale reduction by the energy cascade in the inertial-convective subrange of the turbulent spectrum – wave numbers ranging between the integral and the Kolmogorov scale η (see Fig. 1). Thus, meso-mixing is a homogenization process by advection due to velocity fluctuations. It can be characterized by the TKE k (namely turbulent kinetic energy), a turbulent diffusion concept, or a dominant component of the Reynolds tensor $\overline{u_i u_j}$. In laminar flow, the meso-mixing – on intermediate scales – is related to the possible “mesoscopic” fractal structures of the flow [7].

Micro-mixing is the ultimate mixing scale in the flow; it takes place in the viscous-convective subrange, defined by $\kappa_\eta < \kappa < \kappa_B$ where κ is the wave number, κ_B is the wave number on the Batchelor scale, especially for high-Schmidt-number fluids (liquid phase) when these scales are very different. In this range, turbulent fluctuation has already vanished and mixing is due to laminar stretching proportional to $(\varepsilon/\nu)^{1/2}$, with ε the dissipation rate of the turbulent kinetic energy and ν the kinematic viscosity, that reduces the species clusters on the molecular diffusion scale [8–10]. At the Batchelor scale, the molecular diffusion quickly

dissipates the concentration variance, permitting homogenization of the reagents at molecular scale. The limiting stretching mechanism is related to engulfment in the small vortices near the Kolmogorov scale [4]. Consequently, the micro-mixing depends on the dissipation rate of the turbulent kinetic energy ε , or equivalently on the Kolmogorov scale η .

Actually, if the reaction kinetics is faster than the micro-mixing, the reaction yield is reduced by an insufficient reagent feeding, and the production of by-products is favored; that is why micro-mixing is straightforwardly related to reaction selectivity. In some cases, micro-mixing also affects product quality: for instance, precipitation reaction [11–13] and polymerization reactions [14].

Micro-mixing is responsible for the global performance of multifunctional heat exchanger reactors (MHXR) when the mixing mechanisms on the larger scales – macro and meso-mixing – are not limiting.

Several methods have been reported for characterizing micro-mixing [15]; functional methods fall into three main categories. The first is based on color modification in a solution containing a reacting system directly injected in the flow, such as a pH indicator of an acid-base solution. To obtain quantitative information on mixing efficiency, mixing time is estimated by the ratio of the length of the color variation to the mean flow velocity [16]. The second method involves a spatiotemporal recording of the concentrations to determine a coefficient of variance (COV) and then the segregation index [17]. The third technique, developed by Villermaux [18] and reviewed by Fournier et al. [19] and Guichardon et al. [20,21], entails competitive-consecutive or competitive-parallel reactions that are based on the chemical result of the local injection of a reagent in stoichiometric deficit in the main flow. The main reaction is quasi-instantaneous with a characteristic time t_{r1} ; the side reaction is slower with characteristic time t_{r2} that must be close to the mixing time, so that micro-mixing is limiting for this reaction.

Among these methods, the last-named has been successfully used to evaluate micro-mixing in batch and open-loop reactors with different chemical systems; most of them are consecutive-competing reactions, such as the diazo-coupling test [22], while others are competitive-parallel reactions, such as the iodide/iodate chemical test [19,20,23,24], the method used in the present study.

The principle of the chemical probe is to measure the proportion of the by-products produced due to the secondary slower

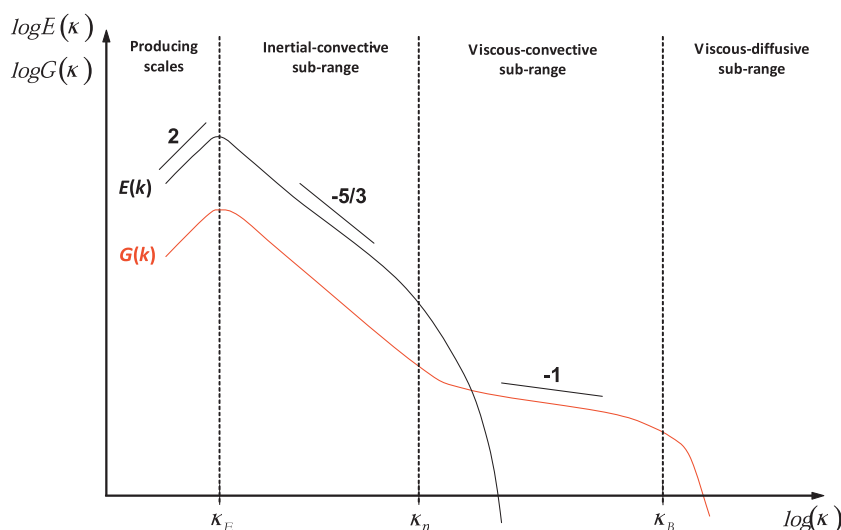


Fig. 1. Spectra of turbulent kinetic energy $E(k)$ and scalar $G(k)$ versus wave number κ (for Schmidt numbers $\gg 1$).

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