INTRODUCTION

The global trend in chemical and manufacturing industries is towards improved energy efficiency, cleaner synthesis, reduced environmental impact and smaller, safer, multifunctional process plants. Such concerns are the driving force for the intensification of batch processes, which are being replaced with continuous high-intensity in-line mass- and heat-transfer equipment. In this context the process intensification (PI) approach, in which the fluid dynamics of the process is matched to the reaction in order to improve selectivity and minimize the byproducts, takes on particular importance.

It is estimated that in a typical large chemical plant, between 5 and 45 million Euros are wasted every year through inefficiencies. In some cases it is estimated that optimization of reactor performance to achieve maximum yield and selectivity would save between 0.4 and 0.8 million Euros per product per year. Overall, reactor-related problems are believed to account for between 0.5 and 3.0% of total turnover, which for European Union chemical industries amounts to 1.9 to 11.4 billion Euros per year.

Systems involving more than one component (or phase) contain interfaces between the components. Our ability to predict the performance of such systems is severely limited. In addition, many multiphase processes are carried out in stirred-tank reactors. Poor flow patterns and low inhomogeneous mixing are characteristic of stirred-tank reactors and typically afford energy dissipation rates in the range 1-10 W.kg⁻¹.

High selectivity requires high rates of micromixing, which need turbulent energy dissipations higher than 100 W.kg⁻¹. Therefore, fast exothermic reactions when carried out in stirred tanks start before mixing is complete, leading to slow apparent rates of reaction and formation of byproducts that must be separated further downline. The high-efficiency vortex (HEV) heat exchanger-reactor, the subject of this study, is selected for its capacity to generate large-scale vortex motions and enhance turbulent energy dissipation in the flow.

A typical example of potential application of this device in manufacturing processes is the ‘mixhead’ of resin transfer molding (RTM) machines. Mixing is often called the heart of RTM and is what differentiates it from other reaction molding processes such as thermoset injection molding or sheet molding. Most mixhead designs were developed by trial and error. Even today, newly designed mixheads are mounted on a machine and a typical (Makosko 1989) reaction is tried on them; mixing quality is then judged by the visual appearance of the product. Therefore, understanding the basic physical phenomena underlying mixing in flows in manufacturing processes is fundamental to a predictive approach to these processes.

In this paper we give a global characterization of a special static mixer designed for use as a reactor-heat exchanger for process intensification in liquid-liquid mixing and reaction. Its design is based on curved tabs fixed on the tube walls that generate longitudinal vortices, substantially increasing transport phenomena over the simple pipe and even over some static mixers known for their high efficiency. Emulsification performance, indicative of the mixing abilities of this system, is presented in this study. Oil-in-water emulsions obtained with the static mixer are characterized by the granulometric distributions. The mean size, size distribution, and power consumption of the mixer are compared with those in some existing devices.

The turbulent characteristics of the flow in the mixer are studied extensively and the physical phenomena underlying the high efficiency of the mixer are addressed.

1. SINGLE-PHASE-FLOW RESULTS

In this section we describe the geometry of HEV, the flow pattern generated and the turbulence properties of the single-phase flow in the mixer.

1.1. Experimental setup and methods

The Perspex HEV test section designed and constructed for this work (figure 1) consists of a straight tube of inner diameter 20 mm along which seven tab arrays are fixed. Each of the seven arrays consists of four trapezoidal tabs positioned at 90° to one another and fixed on the tube walls. The tabs are turbulence promoters and generate longitudinal vortical structures. The test section is 180 mm long and the distance between two successive tab arrays is 20 mm (one tube diameter).

The test section is connected to a preconditioner and postconditioner, which are 300 mm straight transparent tubes of circular inner cross-section and 20 mm inner diameter (the same as that of the test section). The preconditioner is used to provide a fully developed flow at the inlet of the test section, and the postconditioner to observe the effect of the test section on mixing quality.
Injector equipped with a micrometer traversing mechanism

Test section

1st tab array

![Fig. 1: Heat exchanger-reactor HEV.](image)

A schematic diagram of the experimental setup is shown in figure 2. It consists of feed loops for oil and water. The flow rates are controlled by valves and measured with two flowmeters with overlapping ranges.

The working fluid is water and flow patterns are visualized by the laser-induced fluorescence (LIF) technique. Mean and fluctuating velocity fields were measured by laser Doppler velocimetry (LDV) and particle image velocimetry (PIV). Integral length scale and turbulent energy dissipation rates were of particular interest in this work because of their usefulness in modeling the base flow and its effect on droplet breakdown when HEV is used as an emulsifier. Experiments have been run for Reynolds numbers ranging between 1500 and 15000.

![Fig. 2: Experimental rig.](image)

1.2. Single-phase flow results

Figure 3 shows LIF visualizations of the longitudinal vortices downstream of the (a) first, (b) second, (c) third and (d) fourth tab arrays at Reynolds number \(Re = 1500\). At the first tab a pair of vortices is generated that promotes radial mixing. Subsequent tabs increase the degree of mixing, although even at the last tab array special structures can still be observed.

![Downstream 1st tab](image)

![Downstream 2nd tab](image)

![Downstream 3rd tab](image)

![Downstream 4th tab](image)

Fig. 3: Coherent structures at \(Re=1500\).

Figure 4 illustrates the axial mean velocity profiles 2 mm downstream of each tab array for flow \(Re = 15000\) (Mokrani et al. 1995) along axis A. The same figure shows the velocity profile in the straight tube upstream of the first tab array. It can be seen that the first tab array has no significant effect on the mean velocity profile. However, the effect of the second tab array superposed on that of the first induces two minima (behind the tabs) in the mean velocity profile. This deformation of the velocity profile is due to the transfer of low-momentum fluid particles from the HEV wall vicinity to the core flow and is also visible downstream of the subsequent tabs.

![Fig. 4: Global view of evolution of mean axial velocity profile (axis A) at \(Re=15000\) (Mokrani et al. 1995).](image)

Turbulent intensity profiles 2 mm behind each tab array are shown in figure 5 (Mokrani et al. 2002) for Reynolds number 10000 along axis A. The effect of radial flow generated by the streamwise vortices is again visible here; in fact, high-turbulence-intensity fluid particles close to the HEV wall are pushed towards the inner region of the tube, and at the same time the turbulence intensity in the HEV center increases to be more than that at the center of the preconditioner tube.
The above observations suggest that turbulent energy dissipation should also be affected by the vortex generators in the HEV. Turbulent energy dissipation was directly measured by using Batchelor’s energy-cascade model (Batchelor 1953). By using dimensional analysis he modeled the turbulent energy dissipation as:

$$\varepsilon = \frac{3}{2} C \left( \frac{\langle \nu^2 \rangle^{1/2}}{l} \right)$$

(1)

where \(l\) is a length scale in the inertial range. This model predicted well the turbulent energy dissipation in grid-generated turbulence with constant \(C = 1\). However, the validity of the model for highly anisotropic turbulent flows is poor, and the value of \(C\) varies from one experiment to another. Townsend (1976) and Tennekes and Lumley (1997) suggested defining \(\varepsilon\) by the turbulent kinetic energy \(q\):

$$\varepsilon = C \frac{q^{3/2}}{l}$$

(2)

If one assumes that the high-frequency fluctuations involved in the energy dissipation are locally isotropic, and also that the integral macroscale \(A\) can approximate the length scale \(l\), one can define the local turbulent energy dissipation by:

$$\varepsilon = C \frac{\langle \frac{1}{2} u^2 \rangle^{3/2}}{A}$$

(3)

The numerical value of \(C\) must of course be determined experimentally. In this work the integral macroscale \(A\) is determined from the time macroscale by using Taylor’s hypothesis:

$$A = U_{conv} \cdot \sigma$$

(4)

where \(U_{conv}\) represents the convection velocity. This velocity has been determined by using a Van Doorn (1981) expression for three-dimensional flows and simplified through the following hypotheses:

- The axial mean velocity component is much larger than the tangential and radial components,
- The velocity fluctuation field is locally isotropic.

In order to estimate the numerical value of \(C\), a series of experiments was carried out in a straight tube to measure \(\nu^2\) and \(A\). \(\varepsilon/C\) was then calculated from these measured values and compared to the turbulence energy dissipation \(\varepsilon\) experimentally obtained by Lawn (1971) in a straight tube. This comparison allowed determination of the mean value of \(C\), which is fixed at 1.8.

2. LIQUID/LIQUID PHASE: DISPERSION OF OIL IN AN EXTERNAL WATER PHASE

HEV can be used successfully in the emulsification process. Here the focus is on the granulometric characterization of oil-water emulsions obtained in some range of hydrodynamic conditions in the mixer. The droplets are sized using optical microscopy. The mean size, characterized by the Sauter diameter, is fitted to classical turbulent breakup models. The Sauter diameter and the power consumption are compared with those in some existing devices.

2.1. Modeling droplet breakup in turbulent flow

A droplet subjected to mechanical stresses (pressure, velocity gradients or turbulence forces) can burst into several smaller droplets. This phenomenon results from the competitive effects of the rheological and interfacial properties of the two phases and mechanical forces via the flow conditions. For a wide range of applications (stirred vessels, homogenizers, and the like), the breakup is fairly well predicted by a dimensionless Weber number, in formulae that may include the effect of the viscosity of the dispersed phase (the viscosity number). Application of the general breakup model to ‘noncoalesced’ and inviscid systems was tested here following the Shinnar (1961) equation:

$$\frac{d_d}{D} = C_1 \cdot We^{-0.6}$$

(5)

where the Weber number is defined as

$$We = \frac{\rho \cdot U^2 \cdot D}{\sigma}$$

(6)

Among the same lines, Calabrese et al. (1986) justified a corrective term for dilute emulsions to take into account the viscosity of the dispersed phase for applications in agitated tanks. For stirred tanks using Rushton impellers, they recommend:

$$\frac{d_d}{D_{agv}} = 0.054 \left[ 1 + 4.42 \cdot N_a \left( \frac{d_d}{D_{agv}} \right)^{1.5} We^{-0.6} \right] We^{-0.6}$$

(7)

while a similar correlation is proposed by Berkmann and Calabrese (1988) for a Kenics static mixer of pitch 1.5:
\[
\frac{d_{32}}{D_{op}} = 0.49 \left[ 1 + 1.38 N_{vi} \left( \frac{d_{32}}{D_{op}} \right)^{0.5} \right] W^{-0.6}
\]

with viscosity number

\[
N_{vi} = \frac{\mu_{ij} U \left( \frac{\rho}{\sigma} \right)^{0.5}}{d_{32}}
\]

The value of the droplet mean diameter, and thus of the interfacial area, is nevertheless modified by the volume fraction of the dispersed phase from 15 to 30%. The linear corrective term found in Pacek et al. (1998) takes into account the coalescence:

\[
\frac{d_{32}}{D} = C_3 \left( 1 + C_4 \phi \right) W^{-0.6}
\]

### 2.2. Experimental setup and methods

The working fluids are water for the internal phase and an additive technical vaseline oil (Albelf ID 15) for the dispersed phase. The setup allows experiments corresponding to dispersed-phase mixing rates of up to 15%, with Reynolds numbers ranging from 7500 to 15000. A microencapsulation, based on the isocyanate-amine polymerisation reaction, is performed on the emulsion at the exit from the test section.

Table 1. Physical properties of oil loaded with 10% volume of encapsulation reactant.

<table>
<thead>
<tr>
<th>Property (20°C)</th>
<th>Value</th>
<th>Measurement methods</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kinematic viscosity</td>
<td>30.10^{-6} m^2.s^{-1}</td>
<td>Mettler™ RM180 rheometer</td>
</tr>
<tr>
<td>Density</td>
<td>0.85</td>
<td>Technical™</td>
</tr>
<tr>
<td>Interfacial tension with water</td>
<td>20.10^{-3} N.m^{-1}</td>
<td>Krüss™ tensiometer (K12) ring method</td>
</tr>
</tbody>
</table>

A video-optic computer-assisted device was used for granulometric analysis (Pegase software). It was found that a sample of at least 600 droplets is needed to reach 5% precision in the Sauter diameter value.

![Fig. 7: Emulsion samples – optical microscopy.](image)

### 2.3. Results and discussion

Measurements were performed in the 0.1-0.3 global flow rate range, for 0-15% oil volume fraction, in order to investigate the effect of the Weber number on droplet distribution. All experiments were performed at room temperature (18-20°C).

#### 2.3.1. Mean and maximum diameter

Proportionality between the Sauter mean diameter, \(d_{32}\), and the maximum diameter of the drops, \(d_{max}\), is the basic assumption of the turbulent breakup model (Sprow (1967), Pacek et al. (1998)). Experimental results reveal that the diameters \(d_{32}\) and \(d_{max}\) are actually correlated according to the linear relation \(d_{32} = C_3 d_{max}\) (\(C_3=0.48\)).

#### 2.3.2. Sauter diameter as a function of Weber number

The average drop diameter depends strongly on the hydrodynamic conditions through the energy dissipation rate. The HEV measurements in figure 8 show fairly good agreement with this theoretical slope of -0.6 (\(C_2 = 0.57\)), demonstrating that turbulent disruption governs the emulsification mechanism. The fit constant is smaller than 1 (usual value), suggesting that under similar conditions the Sauter diameter is smaller in the HEV than in classical static mixers. This feature will be investigated further by analyzing the energy utilization efficiency of this device.

![Fig. 8: Sauter diameter versus Weber number.](image)

#### 2.3.3. Size distribution function

The method proposed by Schwarz and Bezemer (1956) is applied to fit the present data. The cumulative volume \(V\%) of the distribution function is modeled using a typical constant \(a\):

\[
\ln V\% = \ln 100 + \frac{a - d}{d_{max}}
\]

Figure 9 shows a plot for an emulsion of oil fraction 5%: for each Reynolds number the Schwarz-Bezemer model represents the droplet size distribution in HEV, as far as the straight lines extrapolate to the same intercept.

![Fig. 9: Size distribution function in the Schwarz-Bezemer presentation.](image)

The value of \(a/d_{max}\) is a constant (1.08) independent of the operating conditions, which is more convenient for scale-up operations than a classical log-normal distribution.
3. LOCAL TURBULENCE ANALYSIS

Previous results on the dispersion of oil in water under the combined action of mean and fluctuating velocity fields have raised the complex question of the turbulence parameters relevant in describing the ability of turbulence to split up the oil droplets. In fact, using the local or mean turbulent dissipation rate (measured on axis A) cannot explain the low value of $d_{\text{max}}$. We then made more precise measurements of turbulence (on axis B) in order to explore two different explanations for this discrepancy: first, is it right to consider the turbulence in the HEV as homogeneous and isotropic (equations 3 and 4)? And second, where is the most powerful turbulence for the mixing process located: at the top of the tabs with the highest turbulent energy value, or in their wake where droplets can be trapped and stirred over a long period of time?

3.1. Spatial inhomogeneity of the turbulence

The most significant effect of the tabs is simply to increase the turbulence energy along the tube axis. Figure 10 shows the radial profile of the turbulence energy at three axial positions: $x_u$ upstream of HEV, $x_d$ downstream of the first tab and finally $x_{d7}$ downstream of the seventh one.

![Fig. 10: Radial profiles of the RMS velocity.](image)

If the distribution at $x_u$ corresponds to an equilibrium turbulent flow with turbulence production at the wall, turbulence is completely redistributed inside the HEV, with turbulence production peak at the top of the tabs ($y=0.37R$) that separate the flow in two regions of approximately equivalent turbulent energy: the tube core and the wake of the tabs. Obviously the level of turbulence increases downstream, but Mokrani et al have shown for $Re=10000$ that the turbulence intensity has already reached its maximum value after the second tab.

We then analyzed the quality of these turbulent flows by plotting the probability density functions (pdf) and comparing them with the normal law (see Fig. 11). Figure 11a shows that the pdf in the tube center do not deviate from the normal law either at $x_u$ or at $x_d$, and deviate only transitionally after the first tab. Thus in this region the turbulence is changed not in quality but simply in strength by the action of the tabs. Figure 11b shows that the situation is more complex in the wake region, where the pdf exhibit high skewness and flatness at $x_d$ and $x_{d7}$, indicating that the turbulence there cannot be considered at all homogeneous and isotropic.

![Fig. 11: Probability density functions of the turbulent axial velocity at the three axial positions $x_u$, $x_d$, and $x_{d7}$ in (a) the center and (b) the wake regions.](image)

This schematic description of the flow inside the HEV thus divides the flow into three parts: the turbulent core region with high mean and fluctuating velocities, the wake region with high fluctuating velocity but low mean motion, and the region of sharp shear flow between them. The following sections analyze the turbulence through its spectral energy distribution and its autocorrelation function.

3.2. Turbulence spectrum

To visualize the frequency range of energetic eddies in the turbulence, we plotted the premultiplied spectrum $|\mathcal{E}_{11}|$ with $f$ the frequency and $\mathcal{E}_{11}$ the power density spectrum of the axial velocity component. Then the area under the curve is proportional to the energy of the signal $u'^2$, and can be used to normalize them. Figure 12 compares the normalized spectra at the upstream position $x_u$ with the $-2/3$ power law curve indicating the theoretical energy cascade in the inertial range.

![Fig. 12: Normalized premultiplied spectrum of the turbulent axial velocity in the upstream flow at different radial positions (axis B).](image)

All the curves collapse together with the same integral time scale at the point where the spectra begin to follow the $-2/3$ power law. Then the upstream turbulence inside the tube is quite homogeneous with a simple diffusion of the turbulence energy from the wall to the center and only slight differentiation of the coherent large structure.

On the other hand, spectra at the end of the HEV ($x_{d7}$) are separated into types (figure 13). The most energetic flow is obviously found at the top of the tabs $y/R=0.37$, where the turbulence energy is
produced at higher frequency than in the upstream flow. The integral time scale is roughly ten times smaller than that of \( x_u \).

\[
\begin{align*}
10^{0} & 10^{1} & 10^{2} & 10^{3} & 10^{4} \\
0,00 & 0,05 & 0,10 & 0,15 & 0,20 & 0,25 & 0,30
\end{align*}
\]

\( \text{Frequency, } f \) (Hz)

\( \times u \) wake
\( \times u \) shear flow
\( \times u \) center

**Fig. 13:** Comparison of normalized premultiplied spectra of signals taken at different positions (axis B).

This turbulence diffuses radially and the spectrum at the center is bimodal, with a large-scale similarity to the upstream turbulence and high turbulence level in the inertial range. In the wake region the spectrum is more complex, with a quite large peak at roughly the same integral time scale. Thus, except in this wake region, the turbulence is well modeled by homogeneous and isotropic turbulence.

With the help of these spectra, the integral length scale is calculated by equation 4 and then the local turbulent dissipation rate \( \varepsilon \) is plotted in figure 14 versus \( y/R \) at the three axial positions (even though equations 3 and 4 are not completely valid below \( y/R<0.37 \)). The highest \( \varepsilon \) is found at the high-shear-velocity position at roughly the same level at each tab. This value will be used below to predict the mixing efficiency of this turbulence.

\[
\begin{align*}
\text{Re} = 7500 & \quad \Phi = 0.4 \\
\text{Re} = 10000 & \quad \Phi = 0.5 \\
\text{Re} = 12500 & \quad \Phi = 0.6 \\
\text{Re} = 15000 & \quad \Phi = 0.7
\end{align*}
\]

**Fig. 15:** Ratio of surface energy to available mechanical energy.

The mean energy dissipation rate is computed from the total power consumption for a unit of mass of the processed fluid:

\[
\dot{E} = \frac{Q \Delta P}{\rho V} = \frac{U \Delta P}{\rho L}
\]

Hence the specific energy obtained by integration over the residence time \( L/U \) is:

\[
E = \frac{\Delta P}{\rho}
\]

The pressure-drop values in the test section were measured and compared with the theoretical values for a simple smooth duct of equal hydraulic diameter. The \( z \) factor, defined as the pressure-drop ratio between the HEV and the simple duct,

\[
z = \frac{\Delta P_{\text{HEV}}}{\Delta P_{\text{Duct}}}
\]

as plotted in figure 16, shows that the vortex generators provide up to an eightfold increase in dissipative efficiency, independent of the flow rate.

**Fig. 16:** \( z \) factor.

4. DISCUSSION

In this section we analyze the relations between the granulometric results and the energetic consumption as well as the energy dissipation rate governing the maximum particle diameter, i.e. the breakup process.
Moreover, the pressure-drop measurements show an important decrease from increasing the oil volume concentration, because of the damping of turbulence occurring near the contact surface between the two phases.

4.2 Efficiency
To assess the efficiency of HEV as an emulsifier, its experimentally measured energy cost is compared with those of existing devices as reported by previous investigators. Figure 17 correlates the energy spent in phase dispersion with the interfacial area generated by each system (from Al Taweel et al. 1996). The working zone of the HEV mixer is in the small-energy range (between 0.1 and 1.1 kg/s) for a typical interfacial area of 300-3000 m²/m³. This shows a good efficiency for the dispersion of immiscible liquids: a decrease in energy consumption by a factor of up to 1000 in the range of interfacial areas around 1000 m²/m³.

![Fig. 17: Comparative energy cost of HEV.](image)

4.3 EQUILIBRIUM SIZE
A large amount of laser Doppler velocimetry data was obtained and analyzed in the HEV in order to describe the turbulent energy dissipation distribution in the HEV geometry. Equation 3 was used to evaluate the energy dissipation.

The macroscale \( A \) was derived from the temporal autocorrelation function of the instantaneous velocity. An appropriate convection velocity was calculated by using a generalized Taylor hypothesis. Wu and Patterson (1989) have already obtained satisfactory results using this method.

It is well established that in a given system, where some disruptions of the droplets occur, a maximum equilibrium droplet size is reached that can resist the turbulent forces. It is understandable that the successive divisions leading to this equilibrium size will need a ‘certain’ time. If this time is longer than the residence time, the droplets obtained will be coarser than the predicted size, and thus the system can be optimized by increasing the residence time, for instance by adding another mixing element. The time-scale estimates proposed by some authors (Davies 1985; Hesketh et al. 1991) suggest that the minimal residence time for optimal division is between 1.5 and 10 seconds (the characteristic time for a droplet breakup is estimated at about 1 µs and a sufficient number of divisions must take place).

To analyze these features in the present system, Figure 18 compares the experimental maximum drop size to that predicted by the Hinze theory, using a critical Weber number of order unity:

\[
d_{\text{max}} = \left( \frac{W_{\text{crit}}^2}{2} \right)^{0.6} \left( \frac{\sigma}{\rho_1^{0.4} \rho_2^{0.2}} \right)^{0.6} \epsilon^{-0.4}
\]

The predicted maximum values with the mean turbulence dissipation rate are slightly higher than the measured values. This could mean that the equilibrium diameter has been reached and that the HEV mixer is long enough. Nevertheless, considering the local nature of the dissipation rate and that the maximum ‘surviving’ drop size in the flow is governed by the highest turbulence intensity, the maximum diameter was also computed from the maximum \( \epsilon \) value. From this measure, the equilibrium seems to have been achieved. In other words, it can be concluded from this plot that the actual drop size is controlled by a dissipation rate of the order of the maximum local value. The nonuniformity of the dissipation field may explain the discrepancy between various flow configurations, to the extent that the global behavior cannot be ‘linearly’ deduced from the local analysis.

![Fig. 18: Predicted equilibrium diameter using equation 16.](image)

CONCLUSIONS
The formation of droplets in the turbulent flow of two immiscible fluids in a HEV mixer was studied. The mixing mechanism is by generation of a cascade of longitudinal vortices behind rows of tabs fixed in a straight tube of circular cross-section. By generating extra shear in the flow, vortices increase the turbulent energy dissipation, which contributes directly to droplet breakup.

The very low energy level and the relatively high contact obtained without any surfactant show that HEV mixers have high potential for industrial applications.

Experiments carried out in low hold-up range (0-15%) suggest that the Sauter mean diameter varies with the -0.6 power of the Weber number, demonstrating that the dominant mechanism of emulsification is turbulent disruption.

The size distribution of the droplets obtained with HEV was modeled according to the method proposed by Schwarz and Bezemer (1956). In a log-linear coordinate system the data showed a linear relation between the cumulative volume (in %) of droplet diameters and the inverse of the bounding diameter \( d \), verifying that the Schwarz-Bezemer model applies here.
The overall energy cost of the HEV mixer was also studied. Experimental pressure-drop measurements showed that the vortex generators increase the dissipative efficiency as much as eightfold, and that this increase is independent of the flow rate. The energy consumption for generation of interfacial area with the HEV mixer was compared with those of some common static mixers. In the range of interfacial area $1000 \text{ m}^2\cdot\text{m}^{-3}$, it was shown that HEV is 1000 times more energy-efficient than other mixers.

REFERENCES


Schwarz N. and Bezemer C. 1956. Kolloidzeitschrift 146, 139-145.


