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A TURBULENT APPROACH TO THE DESCRIPTION OF MESO- AND MICROMIXING. APPLICATION TO ACID-BASE REACTIONS IN TUBULAR REACTORS.*

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ABSTRACT

A new approach to the description of turbulence phenomena, based on a different interpretation of the Pao's energy spectrum function, is proposed. An eddy distribution function, that can be used to estimate the characteristic expected value of different turbulence-related properties, is obtained; the contribution of all the eddies in the universal equilibrium range is taken into account. A modified three-stage micromixing model, that can also account for the contribution of macro- and meso-mixing to the total reaction time, is presented. The predictions of the model are compared with experimental results for acid-base neutralisation in a tubular reactor, taken from literature.

INTRODUCTION

If chemical reactions are fast compared to mixing, macro-scale and micro-scale segregation of the reactants may occur. These concentration non-homogeneities influence rate and selectivity of non-first order and complex reactions, affect product properties (e.g. in precipitation and polymerisation) and must be taken into account in scale-up, in the evaluation of reactor stability and in the study of possible thermal run-away.

Mixing is accomplished through the following steps: convection by mean velocity, dispersion by large eddies, reduction of the reactant segregation scale, deformation of the segregated lumps and reduction of the segregation intensity by molecular diffusion.

In the literature these steps are grouped in macro-, meso- and micromixing: a clear distinction between these three mixing modes is questionable; an attempt was proposed by Baldyga and Bourne (1984a) who referred the various steps to different regions of the turbulent concentration spectrum.

The characteristic times of macromixing, τ_{macro}, micromixing, τ_{micro}, and reaction, τ_{kin}, may be evaluated and compared (Ranade and Bourne, 1991; Xi *et al.*, 1991). The great part of the models proposed up to now can deal only the cases in which either macromixing or micromixing is the rate governing process, that is $\tau_{micro} << \tau_{kin} < \tau_{macro}$, or $\tau_{macro} << \tau_{kin} < \tau_{micro}$, but cannot predict the correct performance of the reactor if both macro- and microscale segregation are important (Ranade, 1992). There have been only relatively few efforts of modelling the interaction between the reaction and the whole mixing mechanism (Villermaux, 1989; Baldyga, 1989; Ranade and Bourne, 1991; Thoma *et al.*, 1991; Ranade, 1992; Fox, 1992); a case in which the characteristic times of erosion (dispersive mixing), diffusion and reaction are comparable has been investigated by Klein *et al.* (1980).

In this work a new approach to the description of turbulent effects on mixing is proposed and applied to a three-stage model, that can describe the effects of the different turbulence scales.

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THE NEW APPROACH TO THE DESCRIPTION OF TURBULENT PHENOMENA

In the following, homogeneous isotropic turbulence in an incompressible fluid will be considered. We will move from the three-dimensional energy spectrum function proposed by Pao and Corrsin to obtain an eddy distribution function that can account for viscous dissipation effects out of the inertial subrange.

By means of this function a characteristic turbulent velocity and fluid element deformation will be calculated; their application to modelling of meso- and micromixing will be presented in the next paragraph.

The energy spectrum function $E(k,t)$ may be obtained by the energy balance in the wavenumber space:

$$\frac{\partial E(k,t)}{\partial t} = F(k,t) - 2\nu k^2 E(k,t) \tag{1}$$

where $F(k,t)$ is the energy transfer spectrum function, and the last term on the right-hand side represents the energy dissipation spectrum.
In the universal equilibrium range, the time variation of the energy spectrum function is negligible, and equation (1) may be written (in integrated form):

$$\int_0^k F(k,t)dk + 2\nu \int_k^\infty k^2 E(k,t)dk = 0 \tag{2}$$

The closure hypothesis proposed by Pao (1965) assumes that the total energy flux transferred from the eddies associated to the wavenumber range 0-k through the wavenumber k, which is the first term in the left-hand side of equation (2), is proportional to the energy spectrum function. Accordingly, using dimensional analysis, the following expression is obtained for the energy spectrum:

$$E(k,t) = A\varepsilon^{2/3}k^{-5/3}\exp\left(-\frac{3}{2}A\frac{\nu}{\varepsilon^{1/3}}k^{4/3}\right) \tag{3}$$

By considering that in the inertial subrange, where dissipation by viscous effects is negligible, the well known Kolmogoroff spectrum function is:

$$E_{in}(k,t) = A\ \varepsilon^{2/3}\ k^{-5/3} \tag{4}$$

the energy spectrum function (3) may be written as:

$$E(k) = E_{in}(k,t)\, M(k,t) \tag{5}$$

$M(k,t)$ represents the fraction of the total dissipation rate ε due to the eddies associated to wavenumbers greater than k: introducing the Kolmogoroff microscale $\lambda_K = (\nu^3/\varepsilon)^{1/4}$, it may be written as:

$$M(k,t) = \exp\left(-\frac{3}{2}A\ (k\ \lambda_K)^{4/3}\right) \tag{6}$$

In the following, we will consider stationary turbulence and so we will refer to $M(k)$ only.

In a simplified turbulence representation, in which only the inertial behaviour of the eddies is taken into account, $M(k)$ would be a cumulative distribution function giving the contribution of the eddies associated to wavenumbers higher than k to the spectrum. This is the interpretation that will be utilised in the following; it is more general and more widely applicable than the one previously proposed (Pipino *et al.*, 1992a) and obtained using less stringent hypotheses.

The differential distribution function $R(k)$ may be readily obtained; remembering that according to Tennekes and Lumley (1972) an eddy of wavenumber k is defined as a disturbance containing energy between 0.62k and 1.62k, it follows:

$$R(k) = M(0.62\text{k}) - M(1.62\text{k}) \tag{7}$$

The behaviour of $R(k)$ and $M(k)$ is shown in Figure 1.

The shape of the curves and consequently the position of the maximum of $R(k)$ is slightly influenced by the value adopted for the A constant in equations (3) and (4). The Kolmogoroff spectrum constant is affected by some uncertainty (Hinze, 1975): Tennekes and Lumley (1972, pg. 271) suggest the value 1.5 (and this value was adopted by the authors in previous works and also by Bourne and co-workers); in this work the value 1.7 that allows the best agreement with Pao's experimental data (see Hinze, 1975; pg. 255), will be employed.

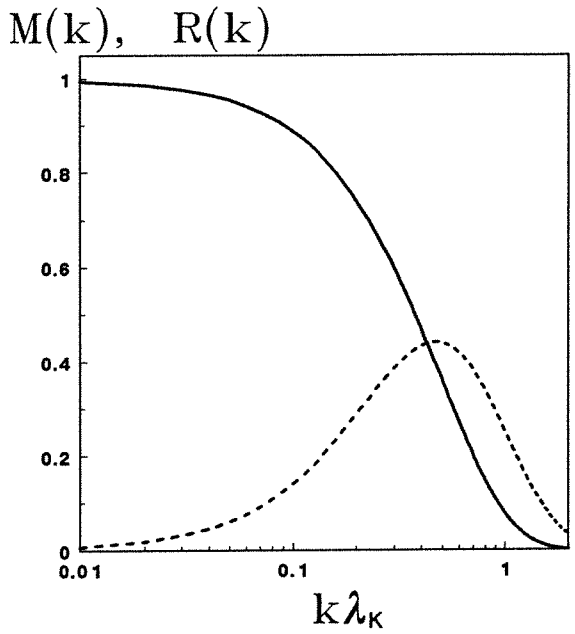

Figure 1. The $M(k)$ [——] and $R(k)$ [-----] functions.

The use of the eddy distribution functions defined above allows a new approach to the description of turbulent phenomena. Every "inertial" quantity or effect that is a linear function of an energy, dependent on the wavenumbers and expressible in the form:

$$g = g(k,t) \tag{8}$$

may be associated to a characteristic value g^*, that takes into account the contribution of the eddies of different scales (Pipino *et al.*, 1992b).

Referring to wavenumbers as integration limits, this characteristic value may be calculated from:

$$g^* = -\int_0^\infty g(k,t)\frac{dM}{dk}dk \tag{9}$$

Characteristic turbulent length and velocity

In what follows, we differentiate between the scales of turbulence and the "physical" properties of the eddies.

We adopt the term "scale" for a quantity related to the spectral analysis of turbulence (for example as defined in Hinze, 1975) and usually derived from dimensional analysis: we will refer to the length scale $\lambda = k^{-1}$ and to the velocity scale $u_s = \varepsilon^{1/3}k^{-1/3}$.

On the other side, if we assign a "physical" size to the eddies associated to wavenumber k, we follow Tennekes and Lumley (1972) in posing $l = 2\pi k^{-1}$ as the characteristic dimension of the eddy.

The "physical" turbulent velocity (i.e. the root mean square of the velocity fluctuations) of the eddies associated to wavenumber k is (Hinze, 1975):

$$u'(k,t) = \sqrt{kE(k,t)} \tag{10}$$

Equation (10) must be consistent with the correlation chosen for l. Following Tennekes and Lumley (1972, pg. 259) in adopting the eddy influence as contained between 0.62k and 1.62k, it is possible to derive for $u'^2(k,t)$ an expression corresponding to equation (10).

From (10) and (4) it follows:

$$u'^2_{in}(k,t) = 1.7\varepsilon^{2/3}k^{-2/3} \tag{11}$$

and applying equation (9):

$$u'^2{*} = -\int_0^\infty u'^2_{in}(k,t)\frac{dM}{dk}dk \tag{12}$$

finally:

$$u'^* = 1.66\ (\nu\varepsilon)^{1/4} \tag{13}$$

This is the characteristic turbulent velocity, calculated taking into account the weighted contribution of all the eddies in the universal equilibrium range.

This characteristic velocity is useful for modelling the convective and distributive mixing stages, and the reduction of the scale of segregation, caused by the largest eddies (with a scale larger than the Kolmogoroff's one). As it will be shown later, this approach allows also the modellisation of the cases in which large scale concentration disuniformities occur.

In the same way, we can obtain a characteristic wavenumber as the one corresponding to the maximum of the $R(k)$ function: it represents the eddy having the greatest importance in the spectrum. We obtained for it the value $13\lambda_K$ which is very close to the value 12 λ_K indicated by Baldyga and Bourne (1984a) as the most effective eddy in the engulfment process.

Deformation of the fluid elements

Baldyga and Bourne (1989) have recently shown that, in many cases, in turbulent liquid media (Sc<4000), diffusion and reaction in deforming laminated structures may not be the rate controlling stage, and do not even affect the product distribution, because the micromixing process is governed by the "engulfment", that is by the rate of formation of the fine laminated structures. On the other side, the previous stages are important when acid-base neutralisations or, in general, almost instantaneous reactions occur.

Several models have been proposed to describe the deformation kinematics (see Hinze, 1975; Ranz, 1979; Ottino *et al.*, 1979; Angst *et al.*, 1982; Bolzern and Bourne, 1983; Baldyga and Bourne, 1984b and Xi *et al.*, 1991), but they all only take into account the effect of the eddies in the viscous subrange. It must be considered that the segregated fluid lumps, even if they are smaller than the Kolmogoroff eddies, may be engulfed and deformed by eddies both in the inertial and in the viscous subrange; moreover, if we assign a "physical" size to the eddies associated to the wavenumber k, it is possible to outline that just the eddies larger than the fluid lump can engulf and deform it.

According to the statistical theory of initial, relative turbulent diffusion (Hinze, 1975), for short diffusion times, the relative diffusion between two fluid points (i.e. their mean square distance s^2) is related to the velocity difference correlation $\mathbf{W}(s_0,t) = \overline{w_i(s_0,t)w_i(s_0,t)}$:

$$s^2 = s_0^{\ 2} + \mathbf{W}(s_0,t)t^2 \tag{14}$$

In order to make a spectral analysis of **W**, we can consider s_0 as the dimension of the eddy associated to wavenumber k, and we can write the velocity difference correlation as a function of wavenumber and time as $W(k,t)$.

Two different correlations can be found for **W** in the inertial and in the viscous subrange.

If $k\lambda_K >> 1$ (viscous subrange):

$$\mathbf{W}(k,t) = \frac{\varepsilon}{3\nu} s_0^{\ 2} \tag{15}$$

and if $k\lambda_K << 1$ (inertial subrange):

$$\mathbf{W}(k,t) = 8.25(\varepsilon s_0)^{2/3} \tag{16}$$

Deformation due to the larger eddies in the inertial subrange is small in comparison of that caused by the viscous subrange eddies, and as a first approximation it has been neglected previously (Barresi *et al.*, 1992), but its contribution may be significative.

The relative importance of viscous and convective effects, in an eddy associated to the wavenumber k, may be estimated by the eddy Reynolds number defined as:

$$\mathrm{Re}_k = \frac{\lambda u_s(k,t)}{\nu} \tag{17}$$

Thus Re_k may be employed in order to build a function able to interpolate the two asymptotic behaviours and describe the velocity correlation in the intermediate range:

$$\mathbf{W}(k,t) = \left(\frac{\varepsilon}{3\nu} s_0^2\right) \frac{1}{1+\mathrm{Re}_k} + (8.25\, \varepsilon^{2/3} s_0^{2/3}) \frac{\mathrm{Re}_k}{1+\mathrm{Re}_k} \tag{18}$$

Applying equation (9), it follows:

$$\frac{\mathbf{W}}{s_0^2} = 0.19 \frac{\varepsilon}{\nu} \tag{19}$$

The variable thickness of the fluid lamella may be calculated as proposed by Baldyga and Bourne (1984b):

$$\left(\frac{d}{d_0}\right)^2 = 1 + \frac{3\mathbf{W}}{2s_0{}^2} t^2 - \left(\left(1 + \frac{3\mathbf{W}}{2s_0{}^2} t^2\right)^2 - 1\right)^{1/2} \tag{20}$$

Substituting equation (19) in equation (20), the rate of shrinkage of the fluid lump is obtained. As expected, equation (19) describes a slower shrinkage than equation (15), that considers all the eddies in the viscous subrange.

EXAMPLES OF APPLICATION TO MESO- AND MICROMIXING MODELLING

Turbulent mixing in a tubular reactor

In a tubular reactor where the two flows containing the reactants A and B are fed separately, coaxially and isokinetically, an highly segregated cone-shaped zone is formed at the outlet of the inner tube (Baldyga and Tovstiga, 1988).

The scale of segregation must be reduced until a mesomixed situation is reached before diffusion and reaction become important.

The characteristic time of this stage, that we will call t_{TM}, "turbulent mixing time", may be readily estimated from the characteristic turbulent velocity u'^* defined by equation (13). If d_i is the diameter of the inner tube, and consequently the initial diameter of the highly segregated B-containing zone, the characteristic time is the one required for the eddies to bring a lump of A-species from the boundary to the centre of the B-rich stream:

$$t_{TM} = \frac{d_i}{2u'^*} \tag{21}$$

and, substituting equation (13) in equation (21):

$$t_{TM} = 0.301 d_i (\nu\varepsilon)^{-1/4} \tag{22}$$

slightly shorter than that previously calculated (Pipino *et al.*, 1992a).

A different formulation of equation (22) may be given, evidencing the influence of the operative parameters. Substituting the expression proposed by Lawn (1971) for the turbulent energy dissipation on the pipe axis:

$$\varepsilon = 4.6 \frac{u_0^3}{D_e} \tag{23}$$

where D_e is the diameter of the external tube and the shear velocity is $u_0 = 0.2\, V \mathrm{Re}^{-1/8}$, we can write:

$$t_{TM} = 0.52 \mathrm{Re}^{-21/32} \frac{D_e d_i}{\nu} \tag{24}$$

The behaviour of equation (24) is shown in Figure 2 (of course the validity is limited to the case $d_i<D_e$). It is interesting to observe that the dependence of t_{TM} on the Reynolds number becomes weaker at higher Re values and with small tubes. Such an observation agrees with the experimental results of Bourne and Tovstiga (1988) and Bolzern *et al.* (1985) where a proportionality between the length of the segregated zone and the diameter d_i of the inner tube (Re = 25,000÷60,000) has been observed.

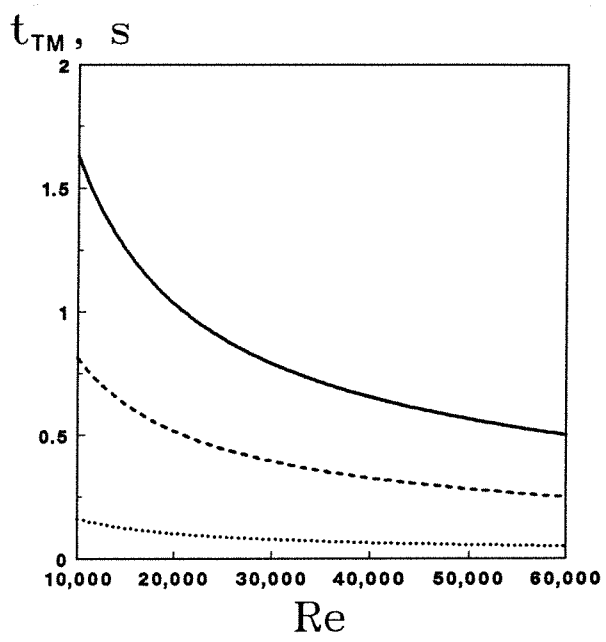

Figure 2. Dependence of the turbulent mixing time on the operating parameters.
$D_e d_i/\nu$ = 1000 [——]; 500 [------]; 100 [·····].

In order to improve the predictions of the model, further informations on the ε field, especially concerning the radial distribution, are needed. In fact, flow disturbances due to the feed tube and additional turbulence generation caused by local velocity gradients may occur, as suggested by the results of Bourne and Maire (1992). Preliminary experimental and computational results evidence that the thickness of the inner tube is also an important parameter, because a wake forms due to the thick trailing edge.

It must be noted that the present model is sensitive to the initial volumetric ratio of the segregated reactants (equation (24) depends on both D_e and d_i), while many models in literature do not have this property (Ranade, 1992). It may be observed that it has some affinities with the engulfment stage of the EDD model proposed by Baldyga and Bourne (1984a), but in that model the characteristic time is the time constant of the eddy with the shortest mean lifetime.

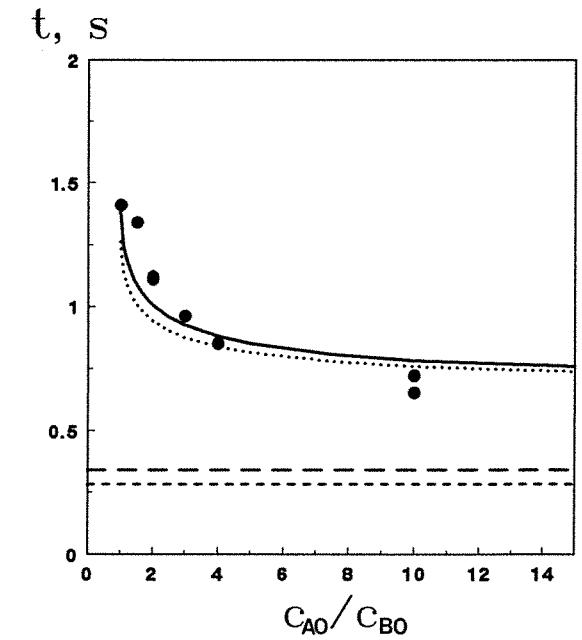

Figure 3. Comparison of the experimental results by Pohorecki and Baldyga (1983) [•] and the predictions of the three-stage micromixing model.
——, this work; ····· Barresi *et al.* (1992).
t_{TM} is also shown: ——, this work; ----, Barresi *et al.* (1992). Re = 13,000; D_e = 0.04; d_i = 5.2·10^{-3} m.

Reaction time of very fast reactions in a tubular reactor

The total reaction time, comprehensive of the mixing time necessary to reduce the scale of segregation below the Kolmogoroff scale, the deformation and the diffusion/reaction time, may be calculated by means of a three-stage micromixing model. The details of the model and its parametric sensitivity have been presented elsewhere (Barresi *et al.*, 1992). In this case the equations for the turbulent mixing time and for the variations of the lamina thickness will be substituted by equations (22) and (20), obtained with the new proposed approach.

The bimolecular, instantaneous, second-order reaction, A+B→P, will be considered and carried out in the tubular

reactor with coaxial feeds previously described. The equations describing diffusion and reaction in the shrinking lamellae, in dimensionless form, are shown in Table 1. The meaning and the advantages of the proposed choice of dimensionless variables have been discussed elsewhere (Barresi *et al.*, 1992)

The predictions of the model may be compared with the experimental results obtained by Pohorecki and Baldyga (1983), who, in an experimental apparatus similar to the one here considered, measured the global mixing-and-reaction time for the NaOH-HCl neutralisation reaction (k_r=10^8 m^3 kmol^{-1} s^{-1} at 293 K)

In Figure 3 the first version of the three stage micromixing model (Barresi *et al.*, 1992) and the modified one, proposed in this work, are compared. A final conversion of 99% has been considered and equal thickness of the lamellae of the two species assumed.

The model is able to predict the experimental results satisfactorily: it has to be evidenced that no best-fitting parameters have been introduced. A better agreement, especially at low reactant concentration ratios, is observed if the modified model using the new turbulent approach is employed.

Table 1. Model equations for the diffusion and reaction stage.

$$\begin{cases} \dfrac{\partial C_A}{\partial T} = \dfrac{1}{\mathrm{Sc}}\left(\dfrac{d_0}{d}\right)_A^2 \dfrac{\partial^2 C_A}{\partial X^2} - \mathrm{M} C_A C_B \\ \dfrac{\partial C_B}{\partial T} = \dfrac{1}{\mathrm{Sc}}\left(\dfrac{d_0}{d}\right)_B^2 \dfrac{\partial^2 C_i}{\partial X^2} - \mathrm{M}\left(\dfrac{c_{A0}}{c_{B0}}\right) C_A C_B \end{cases}$$

$$\left(\frac{d}{d_0}\right)^2 = 1 + 0.285\pi^4 T^2 - \left((1 + 0.285\pi^4 T^2)^2 - 1\right)^{1/2}$$

Initial and boundary conditions:

$T = 0$	$-1 \le X < 0$	$C_A = 0$	$C_B = 1$
$T = 0$	$0 < X \le \xi$	$C_A = 1$	$C_B = 0$
$T \ge 0$	$X = -1, \xi$	$\partial C_i / \partial X = 0$	$(i = \mathrm{A, B})$

Dimensionless variables:

$C_i = c_i / c_{i0}$ $\quad X = 2x/d_B$ $\quad T = (\varepsilon / \nu)^{1/2} \pi^{-2} t$

$\mathrm{M} = (\nu/\varepsilon)^{1/2} \pi^2 c_{B0} k_r$ $\quad \xi = d_A / d_B$

Mixing and reaction in continuous and semi-batch stirred reactors

The previously described model is extensible to the description of fast and complex reactions and single and double jet precipitation in continuous and semi-batch stirred reactors. It may also be easily adopted to handle the slice configuration, that in stirred tank reactors seem to be a more realistic assumption than the lamellar structure (Xi *et al.*, 1991).

In these cases the hydrodynamics is much more complicated, and the variation in the local value of the power dissipation must be taken into account. Work is currently in progress in this direction.

CONCLUSIONS

A new approach to the description of turbulent phenomena has been proposed. It allows the evaluation of the characteristic turbulent velocity, the characteristic eddy size and other turbulence-related quantities, taking into account the contribution of all the eddies in the universal equilibrium range.

By adopting this approach, a modified version of the three-stage micromixing model has been presented. The model has been applied to the description of almost-instantaneous reactions in coaxially fed tubular reactors and is able to handle both meso-scale and micro-scale segregations.

The model is extensible to other reaction systems and reactor configurations, and in particular it may be applied also to continuous and semi-batch tank reactors.

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NOTATION

A	Kolmogoroff spectrum constant, -
C_i	dimensionless concentration of the i-species, -
c_i	concentration of the i-species, kmol m^{-3}
d	lamina thickness, m
D_e	diameter of the outer tube, m
d_i	diameter of the inner tube, m
$E(k,t)$	three-dimensional energy spectrum function, m^3 s^{-2}
$F(k,t)$	three-dimensional transfer spectrum function, m^3 s^{-3}
$g(k,t)$	generic turbulence-dependent quantity,
k	wavenumber, m^{-1}
k_r	reaction kinetic constant, m^3 s^{-1} kmol^{-1}
l	eddy physical size, m
M	modified Damkhöler number, -
$M(k)$	cumulative eddy distribution function, -
$R(k)$	differential eddy distribution function, -
Re	Reynolds number, -
Re_k	eddy Reynolds number, -
s	distance between pairs of fluid points, m
Sc	Schmidt number, -
T	dimensionless time co-ordinate, -
t	time, s
t_{TM}	turbulent mixing time, s
u'	root-mean-square of turbulent velocity, m s^{-1}
$u'(k)$	root-mean-square of the turbulent velocity of the k-associated eddy, m s^{-1}
u_0	shear velocity, m s^{-1}
u_s	turbulent velocity scale, m s^{-1}
V	superficial velocity, m s^{-1}
W	relative velocity correlation, m^2 s^{-2}
w_i	relative velocity of two fluid particles, m s^{-1}
X	dimensionless space co-ordinate, -
x	space co-ordinate, m

Greek letters

ε	turbulent energy dissipation, m^2 s^{-3}
λ	length scale, m
λ_K	Kolmogoroff microscale
ν	kinematic viscosity, m^2 s^{-1}

Superscripts and subscripts

*	characteristic expected value
0	initial conditions
A, B	chemical species
in	inertial formulation

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