

NONIDEAL MIXER AS A SOURCE OF RANDOM FLUCTUATIONS*

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An attempt is made here to describe distribution of residence times of a fluid in a nonideal flow mixer for turbulent flow of the charge, *i.e.* for the case when the flow velocity exhibits random fluctuations. The analysis is based on the assumption that the flow of fluid in mixer may be considered as a stationary Markov's process and is aimed at application of the mixer as a linear filter.

The most frequent approach to study of flow mixer — both from theoretical and experimental point of view — is study of residence time distribution of fluid in this mixer. This approach has been introduced in chemical engineering for the first time probably by Danckwerts¹. Residence time distribution function is used in chemical engineering for analysis of operation of flow units, it is possible to determine the ratio of nonactive (dead) regions and by-pass flow, intensity of backmixing and deviations from ideal mixer and in the case of linear chemical kinetics, conversion in flow reactors².

With development of process modelling by computers attention is paid to another function of the flow mixer which is adequate to the function of a linear filter in communication engineering³. Transfer and first of all smoothing of the input concentration or less frequently of temperature signal by the mixer is concerned. The considered transfer of the signal by flow mixer can be in general described by an integral operator

$$C_e(t) = \int_{-\infty}^{+\infty} \phi(t, \tau) C_i(\tau) d\tau, \quad (1)$$

where C_i and C_e are input or output signals (*e.g.* concentrations) which are in general random functions of time, function ϕ is probability density of residence times in the mixer.

The first studies concerning application of flow mixers for smoothing of non-desirable fluctuations of properties of material streams in continuous processes were published in chemical engineering literature simultaneously with introduction of the concept of residence time distribution in such units. For example Danckwerts¹ and Sellers^{4,5} have in general demonstrated dependence of variance of concentration fluctuations in the outlet stream of the flow mixer on residence time distribution of the liquid in it and on stochastic properties of concentration fluctuations in the input stream of the mixer. Katz⁶ and Kramers with Albeda⁷ have studied smoothing function of

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the cascade of ideal mixers for the case when the input concentration signal is a random stationary signal with the autocorrelation function $R(\tau)$ of the form

$$R(\tau) = \sigma_i^2 \exp(-a|\tau|), \quad (2)$$

where σ_i^2 is the variance of the signal and a is a constant. The same signal at the inlet into the mixer has been considered by Baun and Katz⁸ who have expressed the residence time distribution of liquid in the mixer by χ^2 distribution and *e.g.* by Kraj⁹ and Graichen^{10,11} who have dealt first of all with determination of capacity of the mixer necessary for reaching of the required degree of smoothing of fluctuations.

Considerable attention has been paid in the published studies to smoothing of periodic concentration fluctuations (especially harmonic and rectangular pulses *e.g.*^{12-14,20,21}). These authors have demonstrated that this type of fluctuations can be completely eliminated by suitable arrangement of the mixer (by introducing the by-pass and short-circuiting streams).

Recently there has been published a number of papers¹⁵⁻¹⁹ where an attempt has been made to optimise the smoothing efficiency of the mixer to whose inlet comes a stationary random concentration signal with the autocorrelation function of the form (2) or of a more general form

$$R(\tau) = \sigma_i^2 \exp(-a|\tau|) \cos b\tau, \quad (3)$$

where b is characterising the periodic component present in the input signal, by suitable arrangement of the flow system (*i.e.* by changing the shape of the distribution function of residence times). Conclusions of these studies have demonstrated that by addition of the by-pass (or short circuiting) stream of the mixer it is possible to reach (at suitable selection of parameters of the system mixer-by-pass stream) a better smoothing than with the mixer alone, also in the case of random (not of only periodic) inlet signal.

Very small attention has been devoted in the published literature to experimental determination of smoothing effectiveness of flow mixers and comparison of the so obtained data with theoretical values. Visman and Krevelen²⁰ have measured the degree of smoothing of periodic concentration fluctuations in the bubbled mixer and experimental results were treated in the form of a correlation in which the smoothing degree is plotted against process conditions (volumetric flow rates of liquid and gas, frequency of fluctuations *etc.*) Comparison of experimental data with theoretical model has not been made. Hiby and Tsuge²¹ have measured the degree of smoothing of a periodic concentration signal. The mixer they have used was mixed only by the stream of entering liquid and smoothing degrees measured were in good agreement with values determined on basis of the model of flow in the mixer.

Smoothing action of the flow mixer determine as follows from Eq.(1) two factors - statistical properties of the inlet signal and residence time distribution function of liquid in the mixer. Even that the stochastic character of residence time distribution function in the nonideally mixed flow system has been proved experimentally²²⁻²⁵ and for some cases also described theoretically²⁶⁻²⁹ a very small attention has been devoted in literature to the effect of this fact on the smoothing efficiency of the mixer. For example Václavěk^{24,25} on basis of theoretical analysis by Pugachev³ has used for description of the flow system stationary stochastic operator and has derived a relation describing the increase in value of variance of concentration signal at the outlet from the mixer if compared with the case when the system behaves in deterministic manner. He has demonstrated the decisive role of the autocorrelation function of residence time distribution in the mixer on this increase in variance.

In this study and attempt is made toward explanation of relation between linear operator describing the mixer from the point of view of the transfer of concentration (or some other) signal and hydrodynamic conditions in this nonideally stirred mixer and formulation of assumptions and conditions necessary for description of the smoothing effectiveness of this mixer.

THEORETICAL

BASIC CONCEPTS AND RELATIONS

Let us consider a flow mixer schematically depicted in Fig. 1 of volume Ω_x fully filled by incompressible liquid. Let us assume that it is possible to observe motion of one liquid particle, called indicating particle. Its position in moment t is given by the end of the position vector $\mathbf{X}(t)$. According to the model presented earlier^{30,31} for the one-dimensional space it is considered that the indicating particle is moving so that it is carried in each moment by the liquid with the velocity $\mathbf{V}(t)$. To this macro-motion is superimposed micro-motion due to random interactions with other particles so that the resulting effect is satisfying conditions of the Wiener's process $\mathbf{W}(t)$. Thus the relation holds

$$d\mathbf{X}(t) = \mathbf{V}(t) dt + \sigma d\mathbf{W}(t), \quad (4)$$

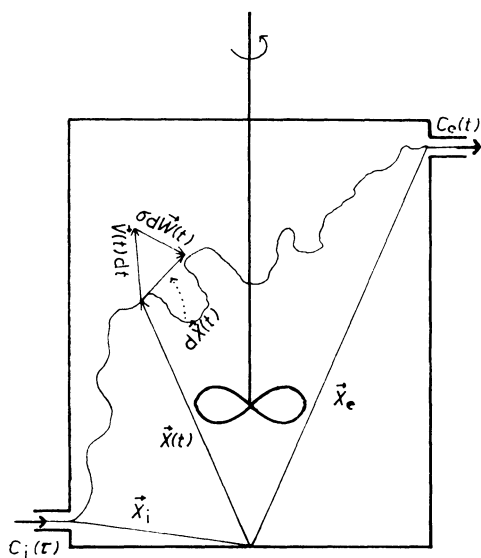


FIG. 1
Motion of indicating particle in the mixer

which is the stochastic differential equation describing motion of the indicating particle (Fig. 1).

Quantities $\mathbf{X}(t)$ and $\mathbf{V}(t)$ are in general random functions of time for which there exists in each moment a probability density function defined by relation

$$f_1(\mathbf{x}, \mathbf{v}; t) = \lim_{\substack{\Delta\Omega_{\mathbf{x}} \rightarrow 0 \\ \Delta\Omega_{\mathbf{v}} \rightarrow 0}} (1/\Delta\Omega_{\mathbf{x}} \Delta\Omega_{\mathbf{v}}) P\{\mathbf{X}(t) \in \Delta\Omega_{\mathbf{x}}(\mathbf{x}), \mathbf{V}(t) \in \Delta\Omega_{\mathbf{v}}(\mathbf{v})\}. \quad (5)$$

The probability that the observed particle is located in the moment t in a small neighbourhood $\Delta\Omega_{\mathbf{x}}$ of the point with the position vector \mathbf{x} is written in the braces. Velocity of liquid and thus also indicating particle velocity in this moment takes value from small neighbourhood of the point \mathbf{v} in the configuration space of velocities $\Omega_{\mathbf{v}}$. Velocity of particle in each moment can be really considered as equal to the velocity of liquid with respect to the fact that the part of change in the position vector $d\mathbf{X}(t)$ corresponding to the Wiener's process does not have a derivative and thus it cannot determine the relative particle velocity with respect to that of liquid^{30,32}. Let us denote that the space $\Omega_{\mathbf{x}}$ of the charge is limited by its boundaries, while the space of velocities is not limited. Relation (5) is not written exactly as there is not prescribed the manner of convergence of small elements $\Delta\Omega_{\mathbf{x}}$ and $\Delta\Omega_{\mathbf{v}}$. More exact definition can be found in corresponding literature (*e.g.*³³). Here the illustrative interpretation of the considered phenomena is considered primarily.

To be able to describe "behaviour" of the particle in the mixer following postulate is accepted: P1. Sum of random and nonrandom effects acting on the liquid (*i.e.* mechanical forces and reactions, first of all action of the mixer) is such that the random position of liquid particles and liquid velocity can be interpreted as stationary Markov process³⁴.

From the made assumption results that forces, either deterministic or of random character could not be explicit functions of time, the mixing unit must be operated so that liquid flow is in steady state.

This assumption makes possible to write the relation for transitive (conditional) probability density for position of particle and liquid velocity in the form

$$\begin{aligned} f_2(\mathbf{x}, \mathbf{v}; t \mid \mathbf{y}, \mathbf{u}; \tau) = \\ = \lim_{\substack{\Delta\Omega_{\mathbf{x}} \rightarrow 0 \\ \Delta\Omega_{\mathbf{v}} \rightarrow 0}} (1/\Delta\Omega_{\mathbf{x}} \Delta\Omega_{\mathbf{v}}) P\{\mathbf{X}(t) \in \Delta\Omega_{\mathbf{x}}(\mathbf{x}), \mathbf{V}(t) \in \Delta\Omega_{\mathbf{v}}(\mathbf{v}) \mid \mathbf{X}(\tau) = \mathbf{y}, \mathbf{V}(\tau) = \mathbf{u}\} \\ [t > \tau], \end{aligned} \quad (6)$$

where the relation in the brace is expressing probability that in time t the particle will be situated in small subspaces $\Delta\Omega_{\mathbf{x}}(\mathbf{x})$ and $\Delta\Omega_{\mathbf{v}}(\mathbf{v})$ at the condition that in preceeding moment τ it has been situated in the point \mathbf{y} and its velocity was equal to \mathbf{u} .

In the case the distribution of particle positions and liquid velocities is known in the moment τ , characterised by the probability density $f_3(\mathbf{y}, \mathbf{u}, \tau)$ it is possible to write

$$f_1(\mathbf{x}, \mathbf{v}; t) = \int_{\Omega_{\mathbf{x}}} \int_{\Omega_{\mathbf{v}}} f_2(\mathbf{x}, \mathbf{v}; t | \mathbf{y}, \mathbf{u}; \tau) f_3(\mathbf{y}, \mathbf{u}; \tau) d\Omega_{\mathbf{v}}(\mathbf{u}) d\Omega_{\mathbf{x}}(\mathbf{y}), \quad (7)$$

where the suggested integration is made, with regard to variables \mathbf{y} and \mathbf{u} over the whole mixer volume $\Omega_{\mathbf{x}}$ and the unlimited configurative space of velocities. Relation (7) is expressing the basic property of the Markov process, *i.e.* that this process in the moment t depends only on the state of the process in a moment $\tau < t$ and is not a function of states in preceeding moments.

For the stationary process the transitive probability density is a function only of the time interval between moments t and τ and not of moments alone³⁴, *i.e.* the relation holds

$$f_2(\mathbf{x}, \mathbf{v}; t | \mathbf{y}, \mathbf{u}; \tau) = f_2(\mathbf{x}, \mathbf{v}; t - \tau | \mathbf{y}, \mathbf{u}). \quad (8)$$

With regard to assumption P1 a stationary probability density for liquid velocity which is not a function of time must exist in each point of the charge, *i.e.* in each point of the space $\Omega_{\mathbf{x}}$. It could be expressed by relation

$$f_5(\mathbf{v} | \mathbf{x}) = \lim_{t \rightarrow \infty} f_4(\mathbf{v}; t | \mathbf{x}). \quad (9)$$

Both these expressions are characterising distribution of liquid velocities in the point $\mathbf{x} \in \Omega_{\mathbf{x}}$ (conditional probability densities are meant) after decay of transition effects, *i.e.* after steading of the process from the moment of beginning of operation of the flow mixer.

Relation between liquid velocities in different time moments from decay of transition effects gives the so called second probability density^{33,34} defined by relation

$$\begin{aligned} & f_6(\mathbf{v}; t - \tau | \mathbf{u}, \mathbf{x}) = \\ & = \lim_{\Delta\Omega_{\mathbf{v}} \rightarrow 0} (1/\Delta\Omega_{\mathbf{v}}) P\{\mathbf{V}(t - \tau) \in \Delta\Omega_{\mathbf{v}}(\mathbf{v}) | \mathbf{V}(0) = \mathbf{u}, \mathbf{X}(t - \tau) = \mathbf{X}(0) = \mathbf{x}\} \quad [t > \tau] \end{aligned} \quad (10)$$

which is with the stationary (so called first) probability density related by relation

$$f_5(\mathbf{v} | \mathbf{x}) = \int_{\Omega_{\mathbf{v}}} f_6(\mathbf{v}; t - \tau | \mathbf{u}, \mathbf{x}) f_5(\mathbf{u} | \mathbf{x}) d\Omega_{\mathbf{v}}(\mathbf{u}) \quad (11)$$

which is expressing the obvious invariability of translation of "steady" distribution of velocities with respect to the time axis.

RESIDENCE TIME OF INDICATING PARTICLE IN THE MIXER

Let us denote motion of indicating particle from the moment of its entry into the mixer up to the moment when this particle leaves the mixer. Further assumption must be introduced in this case: P2. Particle enters into the mixer through the inlet hole in the point \mathbf{x}_i with velocity $\mathbf{V}_i(t)$ which is also a stationary process and leaves the mixer through the exit hole in point \mathbf{x}_e after time interval T with velocity $\mathbf{V}_e(t)$. Each particle can enter and leave the mixer only once.

This assumption first of all simplifies the situation as it substitutes the area of inlet and outlet hole by a single point. This simplification is adequate to the assumption that particle is passing through an arbitrary point of the cross-sectional area of the inlet hole with uniform probability and that distribution of velocities $\mathbf{V}_i(t)$ is also identical for each point of this cross section. Moreover there is assumed that the inequality $\mathbf{V}_i(t) \cdot \mathbf{n}_i < 0$, holds where \mathbf{n}_i is the external normal to the area of inlet hole. Identical assumption holds for the exit hole with the difference that $\mathbf{V}_e(t) \cdot \mathbf{n}_e > 0$.

Finally, this assumption makes possible to write the initial condition for description of motion of the indicating particle in the form

$$f_3(\mathbf{y}, \mathbf{u}; \tau) = \delta_x(\mathbf{y} - \mathbf{x}_i) f_5(\mathbf{u} | \mathbf{y}), \quad (12)$$

where $\delta_x(\cdot)$ is the three-dimensional Dirac function which is expressing the fact that in the initial moment the particle is located in the inlet hole.

Now it is possible to write the probability density $f_1(\cdot)$ for position of the indicating particle and liquid velocity in the mixer which enters into it in moment τ . With respect to Eqs (7), (8) and (12) the relation is obtained

$$f_1(\mathbf{x}, \mathbf{v}; t - \tau) = \int_{\Omega_x} \int_{\Omega_v} f_2(\mathbf{x}, \mathbf{v}; t - \tau | \mathbf{y}, \mathbf{u}) \delta_x(\mathbf{y} - \mathbf{x}_i) f_5(\mathbf{u} | \mathbf{y}) d\Omega_v(\mathbf{u}) d\Omega_x(\mathbf{y}). \quad (13)$$

Let us denote the time interval in this equation by symbol $\theta = t - \tau$ and calculate the marginal probability density for position of indicating particle by integration over the velocity configuration space Ω_v according to relation

$$f_x(\mathbf{x}; \theta) = \int_{\Omega_v} f_1(\mathbf{x}, \mathbf{v}; \theta) d\Omega_v(\mathbf{v}), \quad (14)$$

where $f_x(\cdot)$ is characterising the probability that the indicating particle, in the time interval θ from the moment of entrance into the mixer, will be located in close vicinity of the point \mathbf{x} , without regard to its velocity. Then it is possible to calculate easily the distribution function of residence times in the mixer³³ by use of relation

$$F(\theta) = 1 - P\{T > \theta\} = 1 - \int_{\Omega_x} f_x(\mathbf{x}; \theta) d\Omega_x(\mathbf{x}). \quad (15)$$

The integral in the last term is expressing the probability that the particle is in moment t still in the mixer.

The corresponding probability density for the residence time T is then determined from relation

$$f_7(\theta) = \frac{\partial}{\partial \theta} F(\theta) = - \frac{\partial}{\partial \theta} \int_{\Omega_x} f_x(\mathbf{x}; \theta) d\Omega_x(\mathbf{x}). \quad (16)$$

The function $f_7(\cdot)$ is usually determined experimentally from the mixer response to the input signal in the form of Dirac (unit) impuls. Here is explained why – at least within the limits of the presented model – it is not possible to determine this function by one experiment.

Let us assume first of all that it is possible to interchange the sequence of mathematical operations in Eqs (14) and (16). Then the relation can be written

$$f_8(\theta, \mathbf{v}) = - \frac{\partial}{\partial \theta} \int_{\Omega_x} f_1(\mathbf{x}, \mathbf{v}; \theta) d\Omega_x(\mathbf{x}), \quad (17)$$

which is denoting the joint probability density of particle residence times and liquid velocity independently on position of the particle. This means that expression $f_8(t - \tau, \mathbf{v}) \Delta t$ denotes probability that the indicating particle leaves the mixer in the time interval $[t, t + \Delta t]$ with velocity close to \mathbf{v} (more accurately said by velocity from subspace $\Delta\Omega_v(\mathbf{v})$).

The marginal probability density with respect to variable \mathbf{v} is obtained by integration over all possible values of θ . There obviously holds

$$f_9(\mathbf{v}) = \int_0^\infty f_8(\theta, \mathbf{v}) d\theta = \int_\tau^\infty f_8(t - \tau, \mathbf{v}) dt = \int_{-\infty}^t f_8(t - \tau, \mathbf{v}) d\tau. \quad (18)$$

It is possible to explain by this last equation the physical meaning of function f_9 . The second integral is characterising all possible passages of the indicating particle with various velocities at the assumption that this particle in the moment τ has entered

into the mixer. It is equal to the third integral which at stationary liquid flow is characterising distribution of flow velocities of all liquid particles at the outlet of the mixer in the moment t . Each of these particles could have entered into the mixer in any moment $\tau < t$. Function f_9 is thus characterising distribution of liquid velocities at the outlet from the mixer *i.e.* with respect to Eq. (9) the relation must hold

$$f_9(\mathbf{v}) = f_5(\mathbf{v} \mid \mathbf{x}_c). \quad (19)$$

Let us write finally, probability density for residence times T conditioned with respect to liquid velocity at the outlet from the mixer

$$f_{10}(\theta \mid \mathbf{v}) = \frac{f_8(\theta, \mathbf{v})}{f_9(\mathbf{v})}. \quad (20)$$

Probability density f_{10} is a function of parameter \mathbf{v} which could be randomised³³. According to considerations made earlier³¹ this means that it is possible to consider probability density f_{10} as a function of random liquid velocity $\mathbf{V}_e(\theta)$ at the outlet from the mixer and that it is also a random function of time according to relation

$$f_{10}(\theta \mid \mathbf{v}) = f_{10}(\theta, \mathbf{V}_e(\theta)) = \phi(\theta). \quad (21)$$

Distribution of liquid velocities at the outlet is the result of all, thus also of random effects which act on this liquid during its passage through the mixer. These effects then cause the stochastic character of residence time distributions in the mixer which is expressed by "stochastic" probability density $\phi(\theta)$.

The expected value of this function we can found by integration over the configuration space Ω_v *i.e.* over all values of the randomised parameter \mathbf{v} . With regard to relations (14), (19) and (20) the relation is obtained (again possibility of interchange of the sequence of mathematical operations is proposed)

$$M[\phi(\theta)] = \int_{\Omega_v} f_{10}(\theta \mid \mathbf{v}) f_9(\mathbf{v}) d\Omega(\mathbf{v}) = \int_{\Omega_v} f_8(\theta, \mathbf{v}) d\Omega_v(\mathbf{v}) = f_7(\theta), \quad (22)$$

which is the "usual stochastic" probability density of residence times in the mixer. From Eqs (18), (20) and (21) moreover after integration results

$$\int_0^\infty \phi(\theta) d\theta = \int_0^\infty f_{10}(\theta \mid \mathbf{v}) d\theta = 1, \quad (23)$$

which means that with the probability equal to one the indicating particle leaves the

mixer. The same conclusion also holds for the expected value

$$\int_0^{\infty} f_7(\theta) d\theta = 1. \quad (24)$$

Now the second moment of function ϕ will be calculated.

AUTOCORRELATION FUNCTION FOR PROBABILITY DENSITY OF RESIDENCE TIMES OF INDICATING PARTICLE IN THE MIXER

Second important characteristic for description of motion of the indicating particle is the correlation or autocorrelation function. In the case of Markov processes whose probability characteristics depend at maximum on two time arguments this function is usually sufficient (together with the expected value) for description of the considered process.

Let us define by usual procedure the autocorrelation function for the probability density of residence times ϕ

$$K_{\phi}[\theta, \theta_1] = M[\phi(\theta)\phi(\theta_1)] - M[\phi(\theta)]M[\phi(\theta_1)], \quad (25)$$

where the time interval θ_1 is given by relation $\theta_1 = t_1 - \tau_1$.

Here is explained what is — within the frame of the model presented — the reason for the correlation link between functions $\phi(\theta)$ and $\phi(\theta_1)$. Function $\phi(\theta)$ according to the proposed model is randomised by liquid velocity at the outlet from the mixer $\mathbf{V}_e(\theta)$ and function $\phi(\theta_1)$ by liquid velocity $\mathbf{V}_e(\theta_1)$. Autocorrelation link between these two velocities is obviously the reason of the link between the considered stochastic probability densities of residence time. With regard to Eqs (8), (11) and (19) is the link between velocities at the outlet given by relation

$$f_9(\mathbf{v}) = \int_{\Omega_v} f_6(\mathbf{v}; \theta - \theta_1 | \mathbf{u}, \mathbf{x}_e) f_9(\mathbf{u}) d\Omega_v(\mathbf{u}). \quad (26)$$

Note: According to assumption P2 the parameter \mathbf{x}_e is a constant and will be omitted in the following part.

This approach is now used for definition of the autocorrelation function K_{ϕ}

$$\begin{aligned} K_{\phi}(\theta, \theta_1) = & \int_{\Omega_v} \int_{\Omega_v} f_{10}(\theta | \mathbf{v}) f_{10}(\theta_1 | \mathbf{u}) f_6(\mathbf{v}; \theta - \theta_1 | \mathbf{u}) f_9(\mathbf{u}) d\Omega_v(\mathbf{u}) d\Omega_v(\mathbf{v}) - \\ & - \int_{\Omega_v} f_{10}(\theta | \mathbf{v}) f_9(\mathbf{v}) d\Omega_v(\mathbf{v}) \cdot \int_{\Omega_v} f_{10}(\theta_1 | \mathbf{u}) f_9(\mathbf{u}) d\Omega_v(\mathbf{u}). \end{aligned} \quad (27)$$

It is obvious that autocorrelation function K_ϕ depends not only on intervals θ and θ_1 but also on their difference.

In the case that intervals θ and θ_1 reach the same values, the "randomisation factors", *i.e.* liquid velocities $\mathbf{V}(\theta)$ and $\mathbf{V}(\theta)_1$ will also become identical. Transitive probability density f_6 is so transformed into the Dirac function

$$\lim_{\theta_1 \rightarrow \theta} f(\mathbf{v}; \theta - \theta_1 | \mathbf{u}) = \delta_v(\mathbf{v} - \mathbf{u}) \quad (28)$$

and Eq. (27) after integration with respect to \mathbf{u} becomes into the relation for dispersion of residence time probability density

$$K_\phi(\theta, \theta) = D[\phi(\theta)] = \int_{\Omega_v} f_{10}^2(\theta | \mathbf{v}) f_9(\mathbf{v}) d\Omega_v(\mathbf{v}) - \left[\int_{\Omega_v} f_{10}(\theta | \mathbf{v}) f_9(\mathbf{v}) d\Omega_v(\mathbf{v}) \right]^2. \quad (29)$$

Finally another property of the autocorrelation function $K_\phi(\theta, \theta_1)$ is mentioned. Let us define the integral

$$I = \int_0^\infty [\phi(\theta) - f_7(\theta)] d\theta. \quad (30)$$

The relation for its dispersion holds³

$$D[I] = \int_0^\infty \int_0^\infty K_\phi(\theta, \theta_1) d\theta d\theta_1. \quad (31)$$

With regard to relations (23) and (24) the value of integral I is equal to zero *i.e.* to constant value. Dispersion of constant is then also equal to zero and thus there holds

$$\int_0^\infty \int_0^\infty K_\phi(\theta, \theta_1) d\theta d\theta_1 = 0. \quad (32)$$

Probability characteristics derived previously, *i.e.* the expected value of residence time distribution defined by Eq. (22) and its autocorrelation function, determined by Eqs (25) and (27) with sufficient accuracy — with respect to next considerations — describe random phenomena originating due to the action of the mixing device. These functions can be estimated in the statistical sense by repeated measurements of responses to the unit impulse under the same conditions. The fitness of the estimates increases with the number of measurements repeated.

The relations derived in previous parts are now applied for study of the mixer as a linear filter.

TRANSFORMATION OF RANDOM CONCENTRATION SIGNAL AT PASSAGE THROUGH THE NONIDEAL MIXER

Let us consider a concentration signal of indicating compound $C_i(\tau)$ (Fig. 1) entering continuously into the mixer. An attempt will be made here for determination of the probability characteristics of the outlet concentration signal $C_e(t)$.

In general this relation can be written by use of integral (1) in which the integral core ϕ is in general a random function of two variables (moment of inlet τ and moment of outlet t).

The form of this function or its random character is determined by flow in the mixer, *i.e.* by the action of the mixer and the effect of entering stream. It is demonstrated in which sense this integral core is identical with function ϕ derived in preceding paragraphs, *i.e.* when the equivalence could be written

$$\phi(t, \tau) = \phi(t - \tau) = \phi(\theta) \quad (33)$$

i.e. it is possible to substitute for t and τ their difference.

Further assumption is therefore introduced concerning properties of the inlet signal: P3. Concentration signal $C_i(\tau)$ is a continuous random function of time which is stationary and ergodic with the mean value $\bar{c}_i = \text{const.}$ and autocorrelation function $K_i(h)$. This signal does not depend on liquid distribution entering the mixer and is not affecting by any way flow in the mixer.

Second part of this assumption in accordance with the preceeding considerations implies the statement that the input signal C_i and integral core ϕ are statistically independent. This holds obviously only approximately as fluctuation of liquid velocity in the inlet piping affect to a certain extent input signal. But it is expressing the idea that the basical causes of concentration changes are of different origin (they for *e.g.* originate at production of a component whose concentration is considered) than the effect of flowing liquid, its mixing effect in the inlet piping is thus considered to be negligibly small.

The statement in Eq. (1) is now precised. The assumption of continuous input of the concentration signal makes possible to integrate over the unlimited extent of variable τ . The ergodic property of the input signal will not be considered yet and at first the expected value of both sides of this equation are looked for

$$M[C_e(t)] = M \left[\int_{-\infty}^{+\infty} \phi(t, \tau) C_i(\tau) d\tau \right] = \int_{-\infty}^{+\infty} M[\phi(t, \tau)] M[C_i(\tau)] d\tau. \quad (34)$$

Second relation holds at the assumption that it is possible to interchange the integration with calculation of the expected value and that, as results from assumption P3, the functions ϕ and C_i are independent. With respect to Eq. (22) the expected value of ϕ is a function of the difference of integral variables $\phi = t - \tau$ so that after the interchange of variables the relation holds

$$M[C_e(t)] = \int_{-\infty}^{+\infty} f_7(\theta) M[C_i(t - \theta)] d\theta = \bar{c}_i \int_{-\infty}^{+\infty} f_7(\theta) d\theta = \bar{c}_i. \quad (35)$$

The last equation holds also with respect to Eq. (24). So the familiar relation has been obtained

$$M[C_e(t)] \equiv \bar{c}_e = \bar{c}_i \quad (36)$$

according to which is the expected value of the input concentration signal at considered conditions independent of time and is equal to the expected value of the input signal.

Now the second moment for Eq. (1) is written. With regard to the assumption of independence of functions ϕ and C_i the relation is obtained

$$M[C_e(t) C_e(t_1)] = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} M[\phi(t, \tau) \phi(t_1, \tau_1)] M[C_i(\tau) C_i(\tau_1)] d\tau d\tau_1. \quad (37)$$

With regard to Eqs (25) and (27) the second moment of function ϕ depends only on the difference of their arguments so that after analogous interchange of variables as in calculation of integral (35) the relation is obtained

$$M[C_e(t) C_e(t_1)] = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} M[\phi(\theta) \phi(\theta_1)] M[C_i(t - \theta) C_i(t_1 - \theta_1)] d\theta d\theta_1. \quad (38)$$

Second moments with respect to origin are in this function substituted by central moments, *i.e.* by autocorrelation functions, with Eqs (22), (25), (35) and (36) taken into account the relation follows

$$K_e(t, t_1) + \bar{c}_e^2 = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} [K_\phi(\theta, \theta_1) + f_7(\theta) f_7(\theta_1)] [K_i(t - \theta, t_1 - \theta_1) + \bar{c}_i^2] d\theta d\theta_1. \quad (39)$$

If the subintegral terms in square brackets are multiplied all terms including the constant coefficient \bar{c}_i^2 vanish. First of them because the integral of autocorrelation

function K_ϕ is with regard to Eq. (32) equal to zero. Second term is on basis of Eq. (24) equal to \bar{c}_i^2 , this term vanishes with regard to the second one in Eq. (36) with the corresponding term on the left hand side of relation (39). Finally the autocorrelation function K_i depends according to assumption P3 only on the difference of its arguments. Thus

$$t - t_1 = h \quad (40)$$

and Eq. (39) is simplified

$$K_e(h) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} [f_7(\theta)f_7(\theta_1) + K_\phi(\theta, \theta_1)] \cdot K_i(h + \theta_1 - \theta) d\theta d\theta_1. \quad (41)$$

It is obvious that the autocorrelation function K_e of the output signal is also a function of only a single argument. For identical values of arguments t and t_1 Eq. (41) is reduced to the relation for variance of the output signal

$$K_e(0) = D[C_e(t)] = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} [f_7(\theta)f_7(\theta_1) + K_\phi(\theta, \theta_1)] K_i(\theta_1 - \theta) d\theta d\theta_1 \quad (42)$$

and relation

$$q = \frac{K_e(0)}{K_i(0)} = \frac{D[C_e(t)]}{D[C_i(\tau)]} \quad (43)$$

i.e. ratio of dispersions of the output and input signals is used usually as the measure of smoothing ability of the mixer.

Finally it is demonstrated that at validity of the first part of assumption P3 the characteristics of the output signal can be found from one experimental realisation. The inlet signal is according to this assumption a stationary ergodic function. Value of the autocorrelation function converges to zero if its argument rises to infinity³. Thus there holds $\lim_{h \rightarrow \infty} K_i(h + \theta_1 - \theta) = 0$ and on basis of Eq. (41) also $\lim_{h \rightarrow \infty} K_e(h) = 0$.

The output signal is thus also a stationary ergodic function and for calculation of the mean value and of autocorrelation function the relations can be then used

$$\bar{c}_e = \lim_{T \rightarrow \infty} (1/2T) \int_{-T}^{+T} \int_{-\infty}^{+\infty} f_7(\theta) C_i(t - \theta) d\theta dt \quad (44)$$

$$K_e(h) = \lim_{T \rightarrow \infty} (1/2T) \int_{-T}^{+T} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} [f_7(\theta)f_7(\theta_1) + K_\phi(\theta, \theta_1)] \cdot C_i(h + \theta_1 - \theta + t_1) C_i(t_1) d\theta d\theta_1 dt_1. \quad (45)$$

Relations (43) to (45) can be in practice used for calculation of smoothing effectiveness of the mixer.

DISCUSSION

As has been already stated in introduction of this study, ideas and relations presented here have been aimed at more deep analysis of integral operator in Eq. (1) first of all with regard to smoothing effectiveness of nonideal mixer.

As is known in communication engineering³⁵ or in theory of automatic control this operator is used for calculation of response (in our case denoted as $C_e(t)$) to the input signal $C_i(\tau)$ in systems described by ordinary differential equations (or by their system) with the time argument

$$\sum_{i=0}^n a_i(t) (d^i C_e(t)/dt^i) = C_i(t). \quad (46)$$

It is assumed that the integral core $\phi(t, \tau)$ is a deterministic function of two (time) arguments. Moreover it has been proved³⁵ that as long as the coefficients a_i in the mentioned differential equations are not a function of time both arguments in the integral core can be substituted by their difference, *i.e.* there holds $\phi(t, \tau) = \phi(t - \tau)$. Integral operator then is sometimes called a stationary one. Input signal then might be both deterministic or stochastic with the adequate character of response.

As has been correctly pointed out by Václavek^{24,25} the response can become stochastic even in the case when the input signal is deterministic. This is the case when character of flow in the mixer is causing random fluctuations of the output signal, *i.e.* the integral core ϕ becomes stochastic.

But in this case the coefficients of mentioned differential equations need to be (random) functions of time and the question arises whether or at which conditions it is possible to consider the stochastic integral core as a function of a single argument, *i.e.* whether or at which conditions there exists a stationary stochastic integral operator in the form

$$C_e(t) = \int_{-\infty}^{+\infty} \phi(t - \tau) C_i(\tau) d\tau. \quad (47)$$

Václavek^{24,25} postulates the existence of such operator without proves. This study represents an attempt for a prove of Eq. (47) for the concrete chemical engineering equipment. It is necessary to state that the assumptions made here lead only to the statement that the first two moments of terms in Eq. (47) hold. Analogically to the terminology used in studies of stochastic functions it is possible to state that there exists a stationary stochastic integral operator in a wide sense.

General prove would be perhaps very complicated as *e.g.* it is known³⁶ that, if distribution of the input signal is not Gaussian, it is possible only with difficulties to obtain information on distribution of the input signal. Distribution of the input signal could be obviously approximated by normal distribution in the best case approximately as the concentration signal could reach only nonnegative values. Theoretically it could be possible to use the procedure described in the study of Kattan and Adler³⁷ for computation of conversions of nonideal stirred reactor. But functions of concentration distributions in this study are a priori postulated and it is not obvious how it could be possible to use this procedure in applications.

Relations derived in this study are based on the basic concept of motion of indicating liquid particle in the stationary (in stochastic sense) moving incompressible liquid, while both these processes could be considered to be stationary and Markov. It has been demonstrated earlier^{30,31} that on basis of the law of large numbers motion of large quantity of indicating particles represents a spread of a concentration impuls of the indicating compound. Both these two assumptions are the necessary conditions: Postulate in Markov process property means that motion of particles depends only on initial conditions and is thus not a function of some transitive states in next moments (*i.e.* the familiar analogy exists with deterministic motion of the mass point in the force field). Second assumption on stationarity of the process makes possible to substitute for the two time arguments of the Markov process ("simultaneous" moment t and initial moment τ) by their difference. It is necessary to realize the difference in assumptions on liquid motion and of the indicating compound: It is assumed that while liquid motion has reached steady state before the moment of inlet of the indicating compound (*e.g.* from the beginning of the experiment, when the nonmixed liquid was at rest) concentration of the indicating compound could not steady in the flow system (as long as the input signal depends on time).

To illustrate this statement let us give an example on motion of the indicating particle in an unidimensional unlimited space which could be solved analytically. At assumptions concerning forces which act on this particle made in earlier studies³⁰ it is possible to write the relation for the transitive probability density of particle position and its velocity (illustration of function f_2 in Eqs (6) and (8)) in the form

$$f_2^*(x, v, t - \tau | y, u) = \frac{1}{2\pi (k_{xx}k_{vv} - k_{xv}^2)^{1/2}} \exp \left\{ - \frac{k_{vv}(x - \bar{x})^2 - 2k_{xv}(x - \bar{x})(v - \bar{v}) + k_{xx}(v - \bar{v})^2}{2(k_{xx}k_{vv} - k_{xv}^2)} \right\}, \quad (48)$$

where parameters of this equation are given by relations

$$\bar{x} = \beta(t - \tau) + y + (\beta/\alpha)(\exp(-\alpha(t - \tau)) - 1) + (u/\alpha)(1 - \exp[-\alpha(t - \tau)]);$$

$$\begin{aligned}
 \bar{v} &= \beta + (u - \beta) \exp [-\alpha(t - \tau)], \\
 k_{xx} &= (2\varepsilon^2/\alpha) (t - \tau - [\exp(-2\alpha(t - \tau))/2\alpha] + [2 \exp(-\alpha(t - \tau))/\alpha] - 3/2\alpha), \\
 k_{xv} &= (\varepsilon^2/\alpha) (1 + \exp[-2\alpha(t - \tau)] - 2 \exp[-\alpha(t - \tau)]), \\
 k_{vv} &= \varepsilon^2(1 - \exp(-2\alpha(t - \tau))). \quad (49)
 \end{aligned}$$

Particle velocity equal to the liquid velocity is in this case not a function of its position and corresponding transitive probability density is given by relation (illustration of function f_6 in relation (10))

$$f_6^*(v; t - \tau | u) = \frac{1}{\sqrt{(2\pi k_{vv})}} \exp \left[-\frac{(v - \bar{v})^2}{2k_{vv}} \right]. \quad (50)$$

Relation for steady flow is in this case obtained when the argument t increases to infinity (illustration of function f_5 and of equation (9))

$$f_5^*(v) = \lim_{t \rightarrow \infty} f_6^*(v; t - \tau | u) = \frac{1}{\sqrt{(2\pi) \varepsilon}} \exp \left[-\frac{(v - \beta)^2}{2\varepsilon^2} \right]. \quad (51)$$

It is possible to prove that there holds

$$\begin{aligned}
 f_5^*(v) &= \int_{-\infty}^{+\infty} f_6(v; t - \tau | u) f_5(u) du = \\
 &= \int_{-\infty}^{+\infty} \frac{1}{2\pi\varepsilon \sqrt{(k_{vv})}} \exp \left[-\frac{(v - \bar{v})^2}{2k_{vv}} - \frac{(u - \beta)^2}{2\varepsilon^2} \right] du. \quad (51a)
 \end{aligned}$$

Finally it is possible to use the initial conditions (illustration of function f_3 and relation (12))

$$f_3^*(y, u) = \delta_x(y - x_i) \frac{1}{\sqrt{(2\pi) \varepsilon}} \exp \left[-\frac{(u - \beta)^2}{2\varepsilon^2} \right] \quad (52)$$

and to prove that there holds (illustration of function f_1 and Eq. (13))

$$f_1^*(x, v; t - \tau) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} f_2^*(x, v; t - \tau | y, u) \delta_x(y - x_i) f_5^*(u) du dy, \quad (52a)$$

where the result is also a twodimensional normal distribution as in relation (48)

with the difference that corresponding parameters are given by relations

$$\begin{aligned}\bar{x} &= \beta(t - \tau) \\ \bar{v} &= \beta \\ k_{xx} &= (2\varepsilon^2/\alpha^2)(\alpha(t - \tau) - 1 + \exp(-\alpha(t - \tau))) \\ k_{xv} &= (\varepsilon^2/\alpha)(1 - \exp(-\alpha(t - \tau))) \\ k_{vv} &= \varepsilon^2.\end{aligned}\quad (53)$$

It is obvious that parameter \bar{v} denoting the expected velocity and its variance k_{vv} are not functions of time, other parameters describing motion of particles express, in accordance with the earlier considerations made, spread of concentration of indicating particles in time in the stream of stationary liquid.

Note: It is obvious that if argument t rises particle concentration (proportional to probability density f_1) converges for each point of unlimited axis toward zero. These cases obviously do not have a large practical significance. But in the last study it has been demonstrated³⁸ that in a limited (*i.e.* nonflow) unidimensional mixer it converges to uniform probability density, *i.e.* concentration of indicating particles is of course in steady state at all points of the mixer equal.

Here are also mentioned considerations which lead as limiting cases to usual types of flow in the mixer: From Eq. (4) and on basis of earlier made considerations (for a unidimensional case³⁰) results that there exist two cases of a random shift $d\mathbf{X}$ of the particle in the mixer. This could be either the effect of liquid velocity $\mathbf{V}(t) dt$; in the case this velocity is a random function of time turbulent contribution to random motion is concerned, or in the opposite case laminar flow is concerned. Second term of Eq. (4) $\sigma d\mathbf{W}(t)$ is characterising the diffusion contribution, *i.e.* mutual interactions of particles; second power of coefficient σ^2 is proportional to diffusivity in the normal way. In general case superposition of both contributions takes place.

Let us consider consequences of some simplifications in the proposed model. In the case of laminar, *i.e.* nonrandom flow, dispersion of velocities will be characterised by δ -function, as in the stationary case the liquid velocity is a deterministic function of position (Eq. (9)), given by relation

$$f_5(\mathbf{v} | \mathbf{x}) = \delta_v(\mathbf{v} - \mathbf{v}(\mathbf{x})) \quad (54)$$

or for the output velocity (see Eq. (18)) by relation

$$f_9(\mathbf{v}) = \delta_v(\mathbf{v} - \mathbf{v}_e). \quad (55)$$

On the other hand in the case if diffusion is neglected residence time distribution of a particle is only a function of liquid velocity and conditional probability density

defined by Eqs (20) and (21) will be in this case the Dirac function

$$f_{10}(\theta | \mathbf{v}) = \phi(\theta) = \delta_0(\theta - \bar{\theta}(\mathbf{v})). \quad (56)$$

It is obvious that substitution of the last relation into corresponding equations (Eq. (47)) is expressing the conditions of macromixing. The expected residence time distribution according to Eq. (22) depends only on distribution of liquid velocities; by a slightly different procedure it has been demonstrated earlier³¹ that at certain conditions it is possible to reach the exponential residence time probability density.

Substitution of Eq. (56) into Eq. (29) leads to divergence of the first right hand side integral and variance of residence time distribution at macroflow is thus rising to infinity.

When on the contrary Eq. (55) is substituted into Eq. (29) this variance is equal to zero. This is the case of maximum effect of interaction between individual molecules, *i.e.* the case which according to Zwietering is called the maximum mixedness. Similar considerations have led Hanley and Mischke²² to apply the experimentally found value of variance (more accurately the quantity which is resulting from it) as the measure of "miscibility" in a chemical reactor in which a second order reaction takes place.

Let us consider that interaction between liquid particles is so intensive (diffusivity increases to infinity) that immediately after the entrance of the indicating particle into the mixer is its position characterised by a uniform probability density. This means that this density is not a function of liquid velocity and that its residual residence time does not depend on its preceding history *i.e.* on its age. It is possible to prove that probability density is in this case an exponential function. Variance according to Eq. (29) is obviously equal to zero. Mixer then operates as an ideal one.

When finally Eqs (55) and (56) hold simultaneously the usual piston flow is concerned.

It is also worth mentioning that the mean residence time of a system of indicating particles found at one realisation is a random function and depends on velocity distribution by which is this system carried by the mixer, *i.e.*

$$\bar{\theta}(\mathbf{v}) = \int_0^\infty \theta f_{10}(\theta | \mathbf{v}) d\theta \quad (57)$$

and only the expected value of this quantity gives the familiar relation

$$M[\bar{\theta}(\mathbf{v})] = \int_0^\infty \int_{\Omega_v} \theta f_{10}(\theta | \mathbf{v}) f_9(\mathbf{v}) d\Omega_v(\mathbf{v}) d\theta = \Omega_x / \dot{V}, \quad (58)$$

where \dot{V} is the volumetric flow rate of liquid through the mixer. The analysis made by Václavek²⁴ is not correct in this point, as could be immediately seen after substitution in the extreme case into Eq. (57) from Eq. (56).

But other conclusions reached by Václavek^{24,25} are in full agreement with the relations derived here; it concerns *e.g.* Eqs (23) or (32) which the author has obtained on basis of considerations on the material balance of the input signal. As very important from the point of practical significance is considered Eq. (42). Its first term in square brackets of the subintegral term enables to calculate the "usual" value of dispersion of the output signal at the passage of stationary random signal through the mixer with deterministic properties. Second term represents the stochastic effect of the mixer and is always positive, as has also been proved by Václavek, is equal to dispersion of the integral of product of two centered quantities, *i.e.* of the response to the unit impuls of the mixer and inlet signal and is given by relation

$$\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} K_{\phi}(\theta, \theta_1) K_i(\theta_1 - \theta) d\theta d\theta_1 =$$

$$D \left[\int_0^{\infty} \{ \phi(\theta) - M[\phi(\theta)] \} \{ C_i(\theta) - \bar{c}_i \} d\theta \right] \geq 0. \quad (59)$$

Thus the smoothing ability of the stochastic mixer defined by Eq. (43) is always worse than ability of the deterministic one while this decrease in smoothing ability depends both on function of the own mixer and on stochastic characteristics of the own input signal. Thus it is necessary in designs and calculations of the nonideal mixer as an optimal linear filter always to prove that the proposed system will operate as deterministic one or that stochastic effects of the own mixer are negligible as concerns the smoothing effects.

LIST OF SYMBOLS

| | | |
|----------------------|--------------------------------------|---------------|
| a | constant in Eq. (2) | T^{-1} |
| a_i | coefficients in Eq. (46) | T |
| b | constant in Eq. (3) | T^{-1} |
| c | concentration | ML^{-3} |
| C | random concentration signal | ML^{-3} |
| D | operator of variance | |
| f_1, f_2, f_3 | probability density | $(TL^{-2})^3$ |
| f_4, f_5, f_6, f_9 | probability density | $(TL^{-1})^3$ |
| f_8 | probability density | $(L^{-1})^3$ |
| f_7, f_{10} | residence time probability density | T^{-1} |
| f_x | probability density | $(L^{-1})^3$ |
| h | time interval | T |
| F | residence time distribution function | l |

| | | |
|---------------|--|--------------|
| k_{xx} | variance of position of indicating particle | L^2 |
| k_{xv} | covariance of position and particle velocity | $L^2 T^{-1}$ |
| k_{vv} | variance of particle velocity | $L^2 T^{-2}$ |
| K | autocorrelation function | |
| M | operator of expected value | |
| n | outside normale | 1 |
| P | probability | |
| q | ratio of variances of the output and input signals | 1 |
| R | autocorrelation function | |
| t | time | T |
| T | time interval | T |
| u | vector of initial velocity | LT^{-1} |
| v | velocity vector | LT^{-1} |
| V | random velocity vector | LT^{-1} |
| \dot{V} | volumetric flow rate | $L^3 T^{-1}$ |
| W | Wiener process | $T^{1/2}$ |
| x | position vector | L |
| X | random position vector | L |
| y | initial position vector | L |
| ϕ | residence time probability density | T^{-1} |
| α | parameters in Eqs (49) and (53) | T^{-1} |
| β | | LT^{-1} |
| δ_x | Dirac function | L^{-3} |
| ε | Parameter in Eqs (49) and (53) | LT^{-1} |
| θ | time interval | T |
| σ | standard deviation (see (4)) | $LT^{-1/2}$ |
| τ | time | T |
| Ω | volume | L^3 |

Subscripts

| | |
|-----------|---------------------------------|
| i | assigned to input signal |
| e | assigned to output signal |
| v | assigned to velocities |
| x | assigned to spacial coordinates |
| \bar{c} | mean value of c |

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