ON THE INFLUENCE OF FLUCTUATING FLOW RATE UPON THE PERFORMANCE AND BEHAVIOUR OF ISOTHERMAL REACTING SYSTEMS

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Three simple computational techniques are proposed and employed to demonstrate the effect of fluctuating flow rate of feed on the behaviour and performance of an isothermal, continuous stirred tank reactor (CSTR). A fluidized bed reactor (FBR), in which a non-catalytic gas–solid reaction occurs, is also considered. The influence of amplitude and frequency of gas flow rate fluctuations on reactant concentrations at the exit of the CSTR is shown in four different situations.
Key words: Chemical reactor performance; Fluctuations of input flow rate; Continuous stirred tank reactor; Fluidized bed reactor.

It has been known for two or three decades that intentional unsteady operation of chemical reactors can be superior to the conventional steady-state regime. The advantages sometimes provided by forced process cycling include increased conversion, enhanced selectivity and reduced parameter sensitivity (*e.g.*, refs[1–13]).

In contrast to the above-mentioned forced cycling, fluidized bed reactors exhibit spontaneous oscillations, *e.g.*, in terms of flow rate of gas. The pressure/flow rate fluctuations are closely interrelated with the formation and motion of local inhomogeneities (gas bubbles) within the bed. These fluctuations can be measured by a sensitive pressure transducer, recorded and evaluated by means of statistical analysis (*e.g.*, refs[14,15]). The rising bubbles induce continuous motion of fluidized beds which are normally very well mixed. Therefore, the fluidized bed reactor can often be viewed as a continuous stirred tank reactor (CSTR) in which the concentration and temperature gradients are negligible.

In some our works[16,17], we proposed and verified useful models for sulfur dioxide retention in an isothermal, fluidized bed reactor with sorption of SO_2 by alkaline solid sorbents. These include the conventional steady-state (constant feed flow rate) approach. To our best knowledge, a little has been done towards estimating and exploring the performance of fluidized bed reactors with the fluctuating flow rate of feed in which a non-catalytical gas–solid reaction occurs.

Although the fluidized reactor operates at inherent unsteady state, its design and control are still based on the average values of fluctuating operation quantities. In general, the mean value of the performance of reactor operating at unsteady state will not be the same as the steady-state performance at the mean values of the operation variables. For general aspects of the stochastic modelling of various processes, a reader is referred to the literature (*e.g.*, refs[18–23]).

The use of average values to assess system performance may often be misleading. For example, even though the average temperature can be acceptable, the transient temperature fluctuations may reach values that can cause agglomeration of particles in fluidized beds.

The aim of this work is to propose adequate computational tools which will make it possible to monitor fluidized bed behaviour and reactor performance by simple means as pressure/flow rate fluctuations.

THEORETICAL

Stationary State of Function

Function $f(t)$ will be considered *stationary* for $t > t_0$, if there exists such a quantity P that the average of this function over the interval $\langle t, t + P\rangle$ (sliding average) does not depend on the value of t (*i.e.*, it is constant):

$$\overline{f(t)} = \frac{1}{P}\int_0^P f(t+\tau)\,\mathrm{d}\tau = \text{const.} \qquad (1)$$

If the function $f(t)$ is periodic with the period P, then it is a stationary function. In this work, we consider the function stationarity as equivalent to its periodicity.

Then, a stationary function has the following property: Consider the function $f(t)$ and let it be continuous and differentiable by parts on each interval $\langle t, t + P\rangle$. If we differentiate Eq. (*1*) with respect to t, we obtain

$$(\overline{f(t)})' = \left(\frac{1}{P}\int_0^P f(t+\tau)\,\mathrm{d}\tau\right)' = \frac{1}{P}\int_0^P f'(t+\tau)\,\mathrm{d}\tau = \overline{f'(t)} = \frac{1}{P}\,[f(t+P) - f(t)] = 0\ . \qquad (1a)$$

Analogously to the first derivative of a constant function, the average of the derivative of stationary function over the period P is zero.

Expected Value of Unsteady-State Function as the Average of τ-Realizations of Differential Equation

A differential equation can be written in the general form

$$y'(t) = f(y(t), \boldsymbol{p}(t)) \tag{2}$$

with the initial condition

$$y(0) = y_0 \,, \tag{2a}$$

where $\boldsymbol{p}(t) = (p_1(t), \ldots, p_k(t))$ is the vector of periodic functions with the period P.

By the τ-*realization* of Eq. (*2*), it will be meant the solution of the differential equation

$$y_\tau'(t) = f(y_\tau(t), \boldsymbol{p}(t + \tau)) \tag{3}$$

with the initial condition

$$y_\tau(0) = y_0 \,, \tag{3a}$$

where $\tau \in \langle 0, P \rangle$.

As the *expected* value $\bar{y}(t)$ of solution of Eq. (*3*), we define the average of all τ-realizations over the period P, *i.e.*, the value

$$\bar{y}(t) = \frac{1}{P} \int_0^P y_\tau(t) \, \mathrm{d}\tau \ . \tag{4}$$

By differentiating Eq. (*4*) with respect to t, we get

$$\bar{y}'(t) = \frac{1}{P} \int_0^P y_\tau'(t) \, \mathrm{d}\tau = \overline{y'(t)} \ . \tag{5}$$

As will be shown later, it can be helpful to combine Eqs (*3*) and (*5*) into the form

$$\bar{y}'(t) = \frac{1}{P} \int_0^P f(y_\tau(t), \boldsymbol{p}(t+\tau))\, \mathrm{d}\tau \quad . \tag{6}$$

Let $\lim_{t\to\infty} \bar{y}(t)$ exist. Then, the *expected value* of solution of Eq. (*3*) *at steady state* is given as $\bar{y}_\infty = \lim_{t\to\infty} \bar{y}(t)$. This limit can be estimated with the aid of several techniques as mentioned below.

Outline of Computational Techniques

Method 1

We perform a greater number of τ-realizations of differential equation (*3*) until the time t_0 which can be considered as an onset of stationarity. Upon averaging these results, we obtain the expected value $\bar{y}(t)$ of solution of Eq. (*2*). A certain problem is the estimate of the point t_0. It can be replaced, *e.g.*, by a point which can already be considered as a steady-state solution of Eq. (*2*). However, in this case, the vector $\bar{\boldsymbol{p}} = (\bar{p}_1,\ldots, \bar{p}_k)$ of average (constant) values of functions $p_i(t)$ is inserted into Eq. (*2*) instead of the vector $\boldsymbol{p}(t) = (p_1(t),\ldots, p_k(t))$. As an approximation of steady state of the expected value $\bar{y}_\infty$, we consider then the value $\bar{y}(t_0)$. A drawback of such a procedure is usually a considerable demand on computer time and memory. This method can be viewed as a Monte Carlo method.

Method 2

We perform only one τ-realization of Eq. (*2*) to the time instant t_0, when it is possible to consider the solution stationary in the sense of Eq. (*1*). The estimation of the end point t_0 is carried out analogously to the preceding method. The average value of this τ-realization on the interval $t \in \langle t_0, t_0 + P\rangle$ is then considered as an estimate of the steady-state expected value $\bar{y}_\infty$. We assume that the results for other values $\tau \in \langle 0, P\rangle$ are identical.

Method 3

This method is analogous to the method of computing the steady states in case of the differential equation with constant parameters.

For the differential equation $y'(t) = f(y(t), \boldsymbol{p})$ with constant parameters $\boldsymbol{p}$, we seek the steady-state value y_∞ of $y(t)$ as a solution of the algebraic equation $f(y_\infty, \boldsymbol{p}) = 0$, which follows from the condition $y'(t) = 0$.

In case of the equation $y'(t) = f(y(t), \boldsymbol{p}(t))$, we proceed in the following way.

Solution $y(t)$ will be a stationary function if

$$\overline{y(t)} = \frac{1}{P} \int_0^P y(t+\tau)\, \mathrm{d}\tau = \text{const.} \quad , \tag{6a}$$

$$i.e., \quad \frac{\mathrm{d}\overline{y(t)}}{\mathrm{d}t} = 0 \tag{6b}$$

for all t ∈ ⟨0, ∞).

It also holds

$$\frac{\mathrm{d}\overline{y(t)}}{\mathrm{d}t} = \frac{1}{P}\int_0^P y'(t+\tau)\,\mathrm{d}\tau = [y(t+P) - y(t)] = 0 \ . \tag{6c}$$

When we take $t = 0$ (the initial state), we get the relation $y(P) = y(0)$. At the same time, the function $y(t)$ has to comply with Eq. (*2*).

As a stationary solution, we consider the function given by the solution of Eq. (*2*) with the *boundary* condition

$$y(0) = y(P) \ . \tag{7}$$

The average of solutions of Eqs (*2*) and (*7*) on the interval $t \in \langle 0, P\rangle$ is taken for the expected value of $\bar{y}_\infty$.

In practice, this problem leads to solving a system of algebraic equations. Equation (*2*) can be approximated, *e.g.*, by the Euler implicit difference scheme

$$y'(t) \approx \frac{y(t+\Delta t) - y(t)}{\Delta t} = f(y(t+\Delta t), \boldsymbol{p}(t+\Delta t)) \ , \tag{7a}$$

where $\Delta t = P/n$ and n is a chosen number of mesh points in the interval $\langle 0, P\rangle$. We denote

$$y_i = y(i\,\Delta t)\ , \quad \boldsymbol{p}_i = \boldsymbol{p}(i\,\Delta t) \quad \text{for} \quad i = 1, 2, \dots, n-1$$

and

$$y_0 = y(0)\ , \quad y_n = y(P) \ . \tag{7b}$$

We obtain then a system of $(n + 1)$ algebraic equations for the quantities $y_1, y_2, \dots, y_n$

$$-y_i + y_{i+1} = \Delta t\, f(y_{i+1}, \boldsymbol{p}_{i+1}) \quad \text{for} \quad i = 0, 2, \dots, n-1$$

and

$$y_0 - y_n = 0 \qquad \text{for} \quad i = n \ . \tag{8}$$

After solving this system, we can define the expected value $\overline{y}_\infty$ as

$$\overline{y}_\infty = \frac{1}{n} \sum_{i=1}^{n} y_i \ . \tag{8a}$$

In the preliminary work, all the three methods were thoroughly tested in different situations. The above methods provide identical numerical results in terms of the expected values $\overline{y}_\infty$. Method 3 proved very effective for the estimation of $\overline{y}_\infty$ from the standpoint of needed computer time and memory.

Model of Isothermal, Continuous, Ideally Mixed Reacting System

At constant flow rate of fluid v_0, a simple model of the CSTR at unsteady state is embodied in Eq. (*9*):

$$C'(t) = v_0 \, (C_{\mathrm{in}} - C(t)) - R(C(t)) \qquad \text{for} \quad t \in \langle 0, \infty) \tag{9}$$

with the initial condition

$$C(0) = C_0 \ . \tag{9a}$$

The concentration at steady state C_∞ is given by the solution of the algebraic equation

$$v_0 \, (C_{\mathrm{in}} - C_\infty) - R(C_\infty) = 0 \ . \tag{10}$$

When fluctuations of the flow rate of fluid occur, we can write

$$C'(t) = v(t) \, (C_{\mathrm{in}} - C(t)) - R(C(t)) \qquad \text{for} \quad t \in \langle 0, \infty) \tag{11}$$

with the initial condition

$$C(0) = C_0. \tag{11a}$$

We assume that $v(t)$ is a periodic function with the period P and the average v_0. It is, therefore, a stationary function in the sense of definition given above.

The estimate of the expected steady-state value $\overline{C}_\infty$ can be obtained by any of the methods mentioned above.

Effect of the Relation Between τ_c and τ_v on $\overline{C}_\infty$ at the Reactor Exit

Aside from the effect of amplitude of fluctuations, the behaviour of the solution of Eq. (*11*) depends also on their wavelength τ_v ($\tau_v \approx 1/f$) and on the characteristic response time of the reactor model τ_c. This quantity may be identified *via* linearization of Eq. (*11*) and eigenvalue determination ($\tau_c = | 1/\lambda_c |$). Apparently, two limit situations may be expected:

1) $\tau_v << \tau_c$. Due to its inertia, the solution of Eq. (*11*) does not follow the time changes of fluctuations. It can be approximated by the solution of Eq. (*9*) with the average (constant) flow rate v_0. The expected value of this stationary state C_h will be obtained by solving the algebraic equation $v_0 (C_{in} - C_h) - R(C_h) = 0$.

2) $\tau_v >> \tau_c$. At any time t, it is possible to consider the process as (pseudo-)stable with respect to the instantaneous flow rate $v(t)$. For the expected value of stationary state of this solution C_s, we can then write:

$$C_s = \frac{1}{P} \int_0^P C_s(t) \, dt \ . \tag{11b}$$

The values of function $C_s(t)$ for each $t \in \langle 0, P \rangle$ are given as the solutions of the algebraic equation

$$v(t) \, (C_{in} - C_s(t)) - R(C_s(t)) = 0 \ . \tag{12}$$

In preliminary experiments, a number of model computations were performed with the common reaction term $R(C(t))$ for simple simulated $v(t)$ fluctuations as well as for real (measured by experiment) pressure/flow rate fluctuations. Based on these results, the relation

$$C_s \le \overline{C}_\infty \le C_h \tag{12a}$$

can be viewed as a typical one. $\overline{C}_\infty$ approaches the lower limit C_s (decrease in concentration of the reacted component is coming close to its maximum) for $\tau_v >> \tau_c$. On the other hand, $\overline{C}_\infty$ can attain the upper limit C_h when $\tau_v << \tau_c$. This represents the performance of reactor at the average (constant) flow rate v_0.

Case with a Linear Reaction Term, Correlation Function z(t)

In case of a CSTR with a first-order reaction, it is possible to describe its performance also in another way.

The model takes the form

$$C'(t) = v(t)\,(C_{in} - C(t)) - r\,C(t)\ . \qquad (13)$$

By applying Eq. (*6*), we can obtain, for the expected value of solution $\overline{C}(t)$, the differential equation

$$\frac{d\overline{C}(t)}{dt} = \frac{1}{P}\int_0^P C_{in}\,v(t+\tau) - (r + v(t+\tau))\,C_\tau(t)\,d\tau\ , \qquad (14)$$

where $C_\tau(t)$ is the τ-realization of Eq. (*13*) defined by relation (*3*).

By rearranging Eq. (*14*) we get

$$\overline{C}'(t) = C_{in}\,v_0 - r\,\overline{C}(t) - \frac{1}{P}\int_0^P v(t+\tau)\,C_\tau(t)\,d\tau \qquad (14a)$$

and

$$\overline{C}'(t) = v_0\,(C_{in} - \overline{C}(t)) - r\,\overline{C}(t) - \frac{1}{P}\int_0^P (v(t+\tau) - v_0)\,(C_\tau(t) - \overline{C}(t))\,d\tau\ . \qquad (15)$$

If we denote

$$z(t) = \frac{1}{P}\int_0^P (v(t+\tau) - v_0)\,(C_\tau(t) - \overline{C}(t))\,d\tau \qquad (16)$$

we can finally write

$$\overline{C}'(t) = v_0\,(C_{in} - \overline{C}(t)) - r\,\overline{C}(t) - z(t)\ . \qquad (17)$$

This differential equation gives the expected value of solution $\overline{C}(t)$ of Eq. (*13*).

The function $z(t)$ is a correlation function between the τ-realizations of functions $C_\tau(t)$ and functions $v_\tau(t) = v(t + \tau)$, where $\tau \in \langle 0, P \rangle$. As can be seen, Eqs (*13*) and (*17*) differ only in the term $z(t)$. However, the function $z(t)$ is not *a priori* known. It may be feasible to evaluate it by means of higher moments between its derivatives and the functions $C_\tau(t)$. The differential equation (*17*) can be used in certain considerations on the behaviour of the expected value of solution, especially on its steady-state value $\overline{C}_\infty$.

In this case, the function $z(t)$ has the following properties:

$$z(t) \geq 0 \tag{17a}$$

and converges to the null function ($z(t) \equiv 0$) for $\tau_v \to 0$.

From Eqs (*17*) and (*17a*), the relation $\overline{C}_\infty \leq C_h$ follows, which is part of the above-mentioned Eq. (*12a*).

A typical course of the correlation function $z(t)$ is illustrated in Fig. 1. Different situations with an isothermal CSTR and a fluidized bed reactor (FBR) are summarized in Table I and explored below.

RESULTS

Example 1

Assuming the first-order reaction kinetics for the process of removal of a component out of a fluid, we will estimate dynamic behaviour of the expected value $\overline{C}(t)$ in the sense of the mean value of τ-realizations given by Eq. (*3*).

TABLE I
Summary of computational examples

Example number	Subject of study	Method number	Fluctuations of flow rate of fluid	Reaction kinetics
1	Constant *vs* fluctuating flow rate; $\overline{C}(t)$ as a function of time	1–3	simple oscillatory signal	linear
2	Influence of wavelength (frequency) on $\overline{C}_\infty$	3	simple oscillatory signal	linear
3	Influence of ratio of amplitudes of signals on $\overline{C}_\infty$	3	composite signal of two different oscillations	linear
4	Application to fluidized bed reactor (FBR)	3	real fluctuations measured at FBR in this work	non-linear

The model equation has the form

$$C_\tau'(t) = v(t+\tau)\,(C_{in} - C_\tau(t)) - r\,C_\tau(t) \tag{18}$$

with the initial condition

$$C(0) = C_0 \tag{18a}$$

for $t \in \langle 0, \infty)$ and $\tau \in \langle 0, P \rangle$.

The flow rate of fluid $v(t)$ is assumed to be a periodic function of time with a period P given by

$$v(t) = \frac{p_0}{p(t)}\ , \tag{19}$$

where

$$p(t) = 1 + \alpha \sin(2\pi\, t/\tau_v)\ . \tag{20}$$

We will compute τ-realizations of the solution with a sufficiently large number of mesh points in the τ-interval $\langle 0, P \rangle$ for $t \in \langle 0, t_{max} \rangle$. With respect to the definition (*4*),

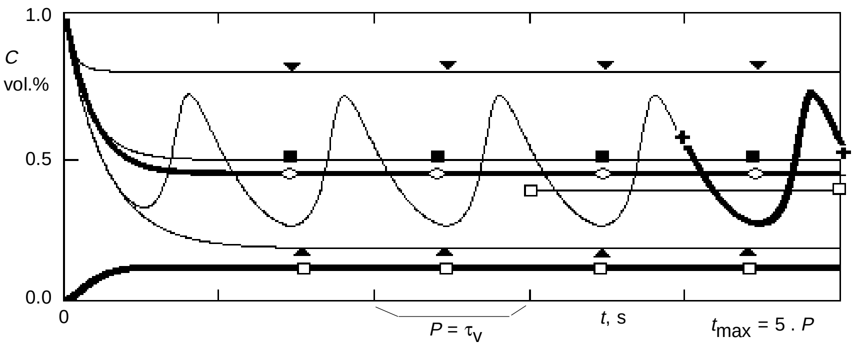

FIG. 1
Expected value of $\overline{C}(t)$ solution evaluated by the method of τ-realizations over the interval $\tau \in \langle 0, P \rangle$ and the correlation function $z(t)$. ✧ Expected value $\overline{C}(t)$, ❒ correlation function $z(t)$, ▼ solution with $v(t) \equiv v_{max}$, ■ solution with $v(t) \equiv v_0$ (average), ▲ solution with $v(t) \equiv v_{min}$, + stationary state, ❒ C_s minimum value ($\tau_v >> \tau_c$), × τ-realization for τ = 0

the expected value $C(t)$ will be taken as the average value of all τ-realizations evaluated at each point t.

The computations were performed for the values as follows: $r = 1\ s^{-1}$, $v_0 = 1\ s^{-1}$, $C_{in} = 1$ vol.%, $P = \tau_v$, $t_{max} = 5P$, $\alpha = 0.9$ kPa and $\tau_v = 0.5$ s. The constant p_0 was taken as large as $p_0 = 0.436$ kPa s^{-1} for which $v(t) = v_0$.

The computed results are plotted in Fig. 1. The courses of the expected value $C(t)$ and the correlation function $z(t)$ defined by Eq. (*16*) are shown in this figure. The results for the constant flow rates ($v_0 = 1\ s^{-1}$, $v_{min} = 0.225\ s^{-1}$ and $v_{max} = 4.36\ s^{-1}$) obtained by a simple τ-realization ($\tau = 0$) are also presented. Furthermore, the stationary state, predicted by Method 3, and the expected value $\overline{C}_\infty$, deduced from it, can also be seen. Finally, the lower bound C_s of evaluated as the mean value of $C_s(t)$, defined implicitly by Eq. (*12*), is shown as well.

The final result can be summarized by the following relation:

$$C_s = 0.390 \text{ vol.\%} < \overline{C}_\infty = 0.446 \text{ vol.\%} < C_h = 0.500 \text{ vol.\%}.$$

It is evident that the expected value $\overline{C}_\infty$ in the stationary state (for the given τ_v) is lower than that for the constant flow rate (C_h). Nevertheless, it is still higher than the lower bound C_s corresponding to the oscillations with $\tau_v >> \tau_c$. As apparent, the size of decrease in the respective concentrations C_s, $\overline{C}_\infty$ and C_s can be viewed as a measure of the reactor performance.

Example 2

Using the same model of CSTR and the same values of its parameters (except for α) as in Example 1, the effect of the wavelength/frequency of flow rate oscillations τ_v on the reactor performance will be explored. In contrast to the above example, we will not seek the expected solution of dynamic behaviour $\overline{C}(t)$, but its steady-state value $\overline{C}_\infty$ evaluated as the average value of a single realization in the stationary state. The stationary solution will be found by Method 3 as mentioned above. The computation will be carried out for values of τ_v which are distributed in a wide range around the characteristic response time τ_c of differential equation (*13*).

The bound values of expected steady state for $\alpha = 0.99$ kPa are as large as

$$C_s = 0.249 \text{ vol.\%} < \overline{C}_\infty < C_h = 0.5 \text{ vol.\%}.$$

The eigenvalue of Eq. (*13*) (λ_c), which is defined as the average value of the eigenvalues $\lambda_c(t)$ at each point $t \in \langle 0, P \rangle$, has the value $\lambda_c = -2\ s^{-1}$ for $\lambda_c(t) \in \langle -15.1\ s^{-1}, -1.07\ s^{-1} \rangle$. As the characteristic response time of the model, we take the quantity $\tau_c = | 1/\lambda_c | = 0.5$ s.

TABLE II
Dependence of the expected value $\overline{C}_\infty$ on the wavelength τ_v of fluctuation, Example 2

τ_v, s	0.02	0.20	2.00	10.0
$\overline{C}_\infty$, vol.%	0.499	0.466	0.266	0.250

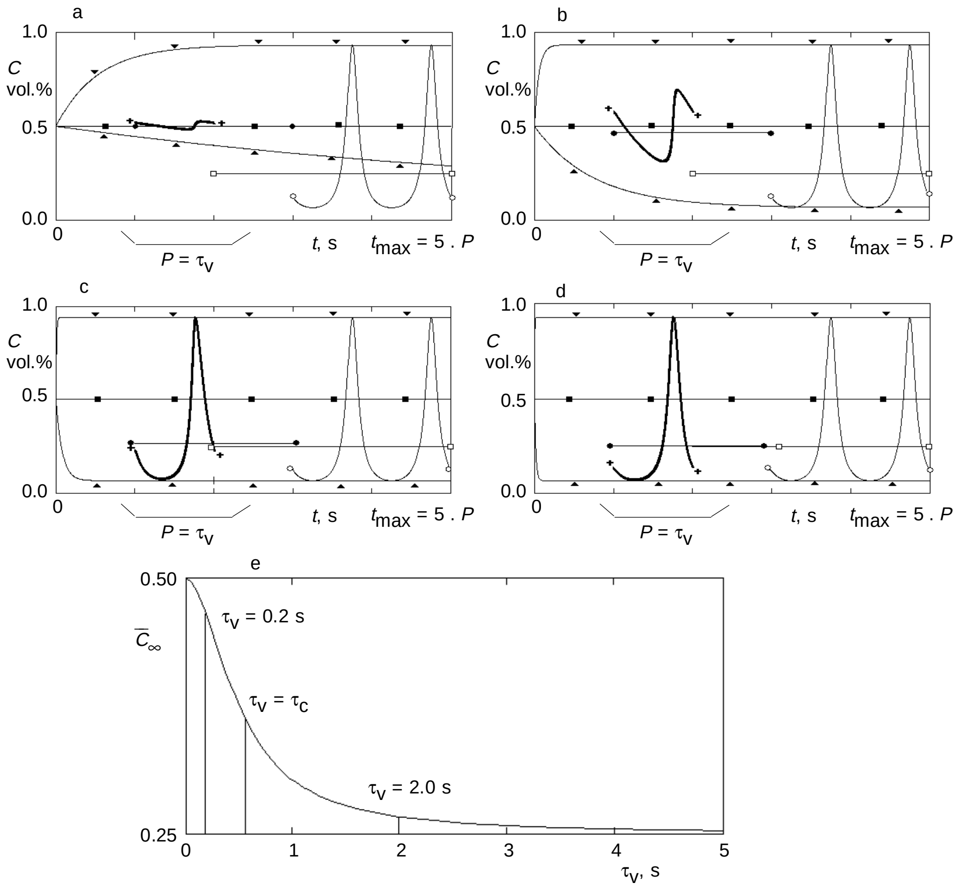

FIG. 2
Effect of wavelength parameter τ_v on the expected value of stationary state $\overline{C}_\infty$ and stationary solution $C(t)$. Solutions for τ_v = 0.02 s (a), τ_v = 0.2 s (b), τ = 2.0 s (c) and τ_v = 10.0 s (d). Diagram e gives the dependence of $\overline{C}_\infty$ on $\tau_v \in \langle 0.01$ s, 5.0 s$\rangle$. ▼ Solution with $v(t) \equiv v_{max}$, ■ solution with $v(t) \equiv \overline{v}_0$ (average), ▲ solution with $v(t) \equiv v_{min}$, + stationary state, ❐ C_s minimum value ($\tau_v >> \tau_c$), ● $\overline{C}_\infty$ expected value of stationary state, ❍ $C_s(t)$ stationary state for $\tau_v >> \tau_c$

The values of $\overline{C}_\infty$, computed for several values of τ_v and shown in Table II, indicate that the reactor performance is higher (decrease in $\overline{C}_\infty$ is deeper) when the wavelength of flow rate oscillations is increased.

All the computed results are plotted in Figs 2a–2d. Solutions are also shown for v_0, v_{min} and v_{max} with the initial condition $C(0) = C_h = 0.5$ vol.% and the stationary state for $\tau_v >> \tau_c$, the mean value of which gives the lower bound C_s. As can be seen in Fig. 2e, the reactor performance (decrease in $\overline{C}_\infty$) is especially sensitive to the changes in wavelength over the interval $\tau_v \in \langle 0.1 \text{ s}, 1 \text{ s}\rangle$.

Example 3

This example illustrates the behaviour of CSTR with two-component oscillations of the flow rate. The respective simple oscillations differ in both frequencies and amplitudes. The oscillations of the flow rate will be assumed in the form

$$v(t) = v_0 \left(1 + \alpha \left[\beta \sin (2\pi\, t/\tau_v) + (1 - \beta) \sin (2\pi\, 10t/\tau_v)\right]\right) \;, \tag{21}$$

where β is the ratio of amplitudes of the respective oscillations. It is apparent from Eq. (*21*) that the frequency of the second component is considered to be ten times higher than that of the first component.

Similarly to Example 2, we will seek only the stationary state of solution and its expected steady-state value $\overline{C}_\infty$. The differential equation

$$C'(t) = v(t)\,(C_{in} - C(t)) - r\,C(t) \tag{22}$$

with the initial condition

$$C(0) = C_0 = 0.5 \text{ vol.\%} \tag{22a}$$

was solved for

$r = 1 \text{ s}^{-1}$, $v_0 = 1 \text{ s}^{-1}$, $C_{in} = 1$ vol.%, $P = \tau_v = 2$ s, $\alpha = 0.9$ kPa and $\beta \in \langle 0, 1\rangle$.

With the use of Method 3, the stationary solutions of Eq. (*22*) and their averages $\overline{C}_\infty$ were evaluated for different values of β. The results of these computations are shown in Figs 3a–3f and in Table III.

TABLE III
Dependence of the values C_s, $\overline{C}_\infty$ and C_h (in vol.%) on the ratio β of amplitude in case of fluctuations a two-component oscilations, Example 3

Parameter	β					
	1.0	0.9	0.7	0.5	0.2	0
C_s	0.425	0.440	0.459	0.464	0.451	0.425
$\overline{C}_\infty$	0.433	0.447	0.470	0.493	0.492	0.492
C_h	0.5	0.5	0.5	0.5	0.5	0.5

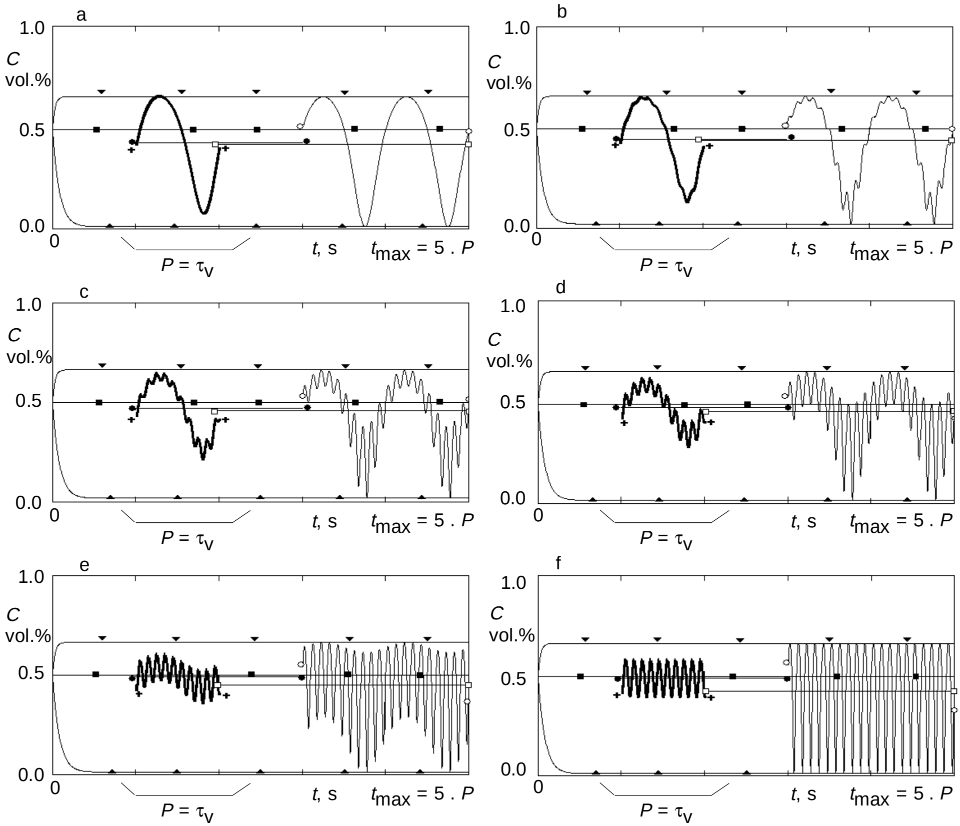

FIG. 3
Stationary state of solution $C(t)$ and expected value $\overline{C}_\infty$ when the flow rate oscillations are composed of two oscillatory components. The ratio of amplitudes β is gradually changed: 1.0 (a), 0.9 (b), 0.7 (c), 0.5 (d), 0.2 (e) and 0 (f). ▼ Solution with $v(t) \equiv v_{max}$, ■ solution with $v(t) \equiv v_0$ (average), ▲ solution with $v(t) \equiv v_{min}$, + stationary state, ❐ C_s minimum value ($\tau_v >> \tau_c$), ● $\overline{C}_\infty$ expected value of stationary state, ❍ $C_s(t)$ stationary state for $\tau_v >> \tau_c$

As can be seen from the obtained results, higher values of β lead to a better performance of the reactor, *i.e.*, to lower values of $\overline{C}_\infty$. The form of Eq. (*21*) suggests that the contribution of the high frequency component decreases as the parameter β increases. In contrast to Examples 1 and 2, the distribution of the flow rates $v(t)$ around the mean value is symmetrical in this Example.

Example 4

In contrast to Examples 1–3, the non-linear reaction kinetics and flow rate fluctuations monitored in this work during the operation of a fluidized bed reactor are employed below. Unsteady-state sorption of sulfur dioxide by magnesium oxide in a fluidized bed[16] can be described as

$$C'(t) = v_0\,(C_{in} - C(t)) - \frac{t_s}{t_g}\,C_{in}\,R(C,X)\ , \tag{23}$$

where

$$R(C,X) = X'(t) = 0.365\,C^{0.873}\,(0.4 - X) \tag{24}$$

and the initial conditions are $C(0) = C_0 = C_{in} = 0.33$ vol.% and $X(0) = 0$.

The parameters $\overline{t}_g$ and t_s are greatly different (0.23 s and 2 . 10^4 s, respectively), which indicates that the time scales of the concentration changes in the gas and solid phase are very different.

The constant flow rate v_0 was replaced by the fluctuating function $v(t)$ with the mean value v_0. An experimental sample was employed with 128 readings of $v(t)$ measured in a fluidized bed reactor during $P = 19.2$ s of its operation. The fluctuating flow rate $v(t)$ is shown in Fig. 4a. In this Example, $v(t)$ fluctuated between $v_{min} = 0.2\ s^{-1}$ and $v_{max} = 8.4\ s^{-1}$ with $v_0 = 4.3\ s^{-1}$. Since this period of time is short, the conversion of the solid sorbent was assumed to be time independent and equal to $X = 0.3$.

Using Method 3, the stationary solution $C(t)$ was found from which the expected value $\overline{C}_\infty$ was deduced. The computed stationary state is shown in Fig. 4b.

To explore the effect of frequencies on $\overline{C}_\infty$, computations were also performed for $P = 0.2$ s, *i.e.*, the frequencies of the fluctuating velocity $v(t)$ were increased by a factor of 19.2/0.2 = 96. The results of the two computations are presented in Table IV.

As can be seen, the much higher frequencies of the flow rate fluctuations lead to a higher value of $\overline{C}_\infty$, *i.e.* to somewhat lower performance of the fluidized bed reactor for SO_2 removal. This finding is consistent with the results in Examples 2 and 3 and is in general agreement with the conclusions of other authors[12].

CONCLUSIONS

Three employed computational techniques provide practically identical results. Methods 1 and 2 are more general, however, their applications usually need more computer time. It should be noted that Method 1 makes it possible to estimate the expected output quantity (*i.e.*, $\overline{C}_\infty$) in the course of time.

Method 3 is simple and rapid. This procedure is particularly useful when τ_v and τ_c differ in the order of magnitude.

We believe that these computational tools will also be useful in analyzing other systems with the fluctuating quantities.

TABLE IV
Values of C_s, $\overline{C}_\infty$ and C_h (in vol.%) at different frequencies, Example 4

P, s	C_s	$\overline{C}_\infty$	C_h
19.0	0.143	0.144	0.158
0.2	0.143	0.153	0.158

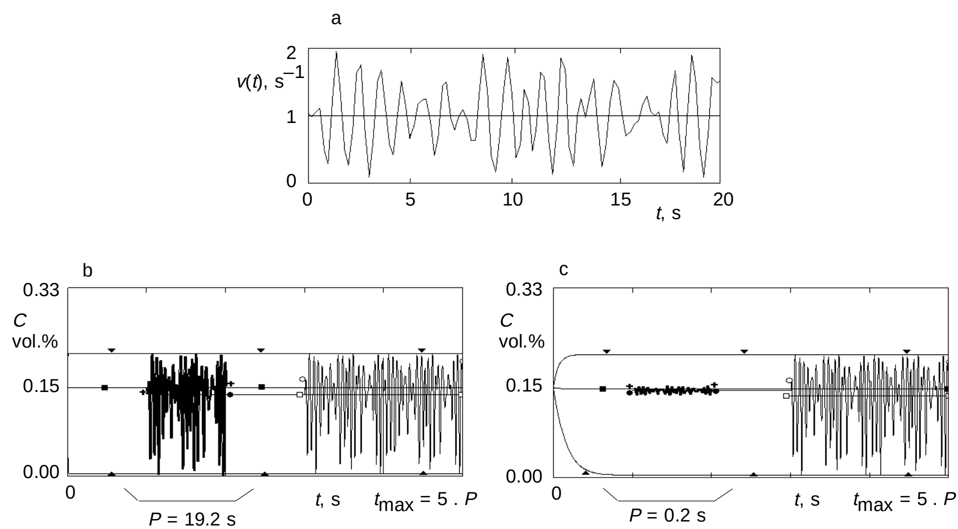

FIG. 4
The solutions of Eqs (*23*) and (*24*) modelling the sulfur dioxide removal in a semi-continuous fluidized bed reactor. Measured flow rate fluctuations, shown in diagram a, are employed. Diagram b shows the stationary solution $C(t)$ and the expected value $\overline{C}_\infty$ for the measured P = 19.2 s. Diagram c depicts $C(t)$ and $\overline{C}_\infty$ for P = 0.2 s. ▼ Solution with $v(t) \equiv v_{max}$, ■ solution with $v(t) \equiv v_0$ (average), ▲ solution with $v(t) \equiv v_{min}$, + stationary state, ❐ C_s minimum value ($\tau_v >> \tau_c$), ● $\overline{C}_\infty$ expected value of stationary state, ❍ $C_s(t)$ stationary state for $\tau_v >> \tau_c$

It has been confirmed that the slow fluctuations of input flow rate with larger amplitudes appear to be favourable for the performance of a reactor system.

SYMBOLS

$C(t)$	concentration in reactor at time t, vol.%
$\overline{C}(t)$	expected (average) concentration at time t, vol.%
C_{in}	concentration at inlet of reactor, vol.%
C_h	stationary state of concentration at constant flow rate v_0, vol.%
C_s	average value of $C_s(t)$ on interval $t \in \langle 0, P \rangle$, vol.%
$C_s(t)$	pseudo-stationary state at time $t \in \langle 0, P \rangle$, solution of equation $v(t)\,(C_{in} - C_s(t)) - R(C_s(t)) = 0$, vol.%
$C_\tau(t)$	τ-realization of differential equation for fluctuating flow rate, vol.%
C_0	initial concentration at time $t = 0$, vol.%
C_∞	stationary state of concentration at constant flow rate v_0, vol.%
$\overline{C}_\infty$	expected (average) value of function $\overline{C}(t)$ at steady state, vol.%
f	frequency of fluctuations, s^{-1}
n	number of mesh points in the interval $\langle 0, P \rangle$
p_0	constant in Eq. (*19*), kPa s^{-1}
$p(t)$	pressure as function of time, kPa
P	period of periodic function, s
$\boldsymbol{p}$	vector of constants
$\boldsymbol{p}(t)$	vector of functions dependent on time
r	reaction rate constant, s^{-1}
$R(C)$	reaction term, s^{-1}
t	time, s
t_{max}	maximum time, s
t_{min}	minimum time, s
t_0	instant from which function is considered stationary in the sense of Eq. (*1*), s
$\overline{t}_g$	mean residence time of gas in fluidized bed, s
t_s	stoichiometric time for semi-continuous fluidized bed reactor (minimum period of time needed for complete conversion of sorbent in the bed), s
v_0	average (constant) flow rate of fluid, space velocity, s^{-1}
$v(t)$	instantaneous (fluctuating) flow rate of fluid, space velocity, s^{-1}
v_{max}	maximum flow rate of fluid, space velocity, s^{-1}
v_{min}	minimum flow rate of fluid, space velocity, s^{-1}
$X(t)$	conversion of solid sorbent at time t
$\overline{y}(t)$	expected (average) value of τ-realizations of differential equation over interval $\tau \in \langle 0, P \rangle$
$y_\tau(t)$	τ-realization of differential equation
$\overline{y}_\infty$	expected (average) value of steady state of function $\overline{y}(t)$
$z(t)$	correlation function between flow rate $v(t)$ and solution $C(t)$ defined by Eq. (*16*), s^{-1}
α	amplitude of fluctuation signal, kPa
β	ratio of amplitudes of oscillations in composite signal in Eq. (*21*)
λ_c	eigenvalue of model equation, s^{-1}
τ	parameter of τ-realization of differential equation, s
τ_c	characteristic response time, $\tau_c = \vert 1/\lambda_c \vert$, s
τ_v	wavelength of periodic sample of fluctuations, s

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