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**CERTAIN PROBLEMS WITH THE APPLICATION OF STOCHASTIC  
DIFFUSION PROCESSES FOR DESCRIPTION OF CHEMICAL  
ENGINEERING PHENOMENA; MODELLING OF PROCESSES  
BY MEANS OF STOCHASTIC DIFFERENTIAL EQUATIONS**

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Accepted October 16th, 1987

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The paper points at certain problems associated with direct use of stochastic differential equations for description of chemical engineering processes or with the use of corresponding diffusion equations. It is shown that on the basis of various definitions one can write down three types of stochastic differential equations which might, in principle, describe the same process. One of these types is at the same time equivalent to the classic transport equations common in chemical engineering. A method is described removing these inconsistencies.

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The previous communication<sup>1</sup> pointed at the differences in the notation of transport equations employed in chemical engineering, i.e. differential mass balances of the component or energy balances in the flowing fluid and the notation of the Kolmogorov diffusion equations, derived in the mathematical theory of random processes (see e.g.<sup>2,3</sup>).

Specialized mathematical literature<sup>2,3</sup> further proves that these equations are associated with the stochastic differential equations enabling direct description of the development of a random process in time. This fact has been used in chemical engineering, as well as in many other disciplines, in such a way that the stochastic differential equations permit one to formulate relatively simply the physical model of a chemical engineering random process<sup>4</sup>. This relationship can be further modified by using simple rules to obtain the Kolmogorov parabolic differential equation for the complete probability characteristic of the process — its distribution function or the probability density function. Solution of this last equation enables us to obtain a detailed information about the process; the probability density may then be often regarded to be a function proportional to the concentration of the component or the temperature of the medium.

There exist a large number of papers applying this procedure; some of them have been quoted in the preceding communication<sup>1</sup>. Here we shall therefore quote only reviews<sup>5-7</sup> and chemical engineering monographs<sup>4-8</sup> explaining the application of such a process.

Recently, in connection with the development of powerful computers, there is a tendency to model processes directly by means of stochastic differential equations; in chemical engineering this approach is being applied, for instance, for description of chemical reactors<sup>9-11</sup>. A broader exploitation of this method, however, is being hampered, according to our opinion, by the fact that in the specialized mathematical literature two methods are used for notation of the stochastic differential equations – the Ito and the Stratonovich notation. These two notations may, in some cases, provide from the viewpoint of practical application different results (see e.g. ref.<sup>4</sup>).

From the theoretical viewpoint this dual character of the notation is determined by two ways of defining the stochastic integral<sup>4,12,13</sup>. In the following paragraph we shall therefore briefly point out these differences. We shall show that one can set up another definition leading to a third way of notation of the stochastic differential equations and that this approach is sometimes adequate to the “classic” transport equations used in chemical engineering.

## THEORETICAL

### *Description of Motion of a Particle in the Flowing Fluid*

Equally as in the preceding communication<sup>1</sup> we shall aim at illustrative concepts and applications of the mathematical model considered to a concrete process: the motion of a particle of a distinguishable component in the flowing fluid (in a less illustrative case the motion of an energy quantum in this fluid) taking place in the three-dimensional Euclid space. This particle is taken to be a mass point and its position shall be determined by the position vector  $\mathbf{X}(t)$ . Let us assume that the particle, on the one hand, is carried away together with the fluid and, on the other hand, moves also relatively to the fluid, this latter motion being caused by random collisions with particles of the fluid surrounding it. The effect of the external forces is therefore regarded as insignificant.

The situation considered may be described by a relatively simple kinematic model expressing the velocity of the distinguishable particle as a linear superposition of the velocity of the fluid  $\mathbf{v}$ , carrying the particle and the relative velocity of the particle with respect to the fluid:

$$d\mathbf{X}(t)/dt = \mathbf{v}(\mathbf{X}(t), t) + \mathbf{G}(\mathbf{X}(t), t) \cdot \xi(t). \quad (1)$$

In this equation the velocity  $\mathbf{v}$  is generally considered to be a deterministic function of the instantaneous position of the particle and explicitly also time. The first coefficient of the last term is generally a second order tensor and also a deterministic function of the position and explicit function of time. It characterises also the properties of the medium in which the particle moves and if this medium is homogeneous, isotropic and does not vary in time, it is merely a scalar constant.

The second coefficient  $\xi(t)$  determines the random character of the interactions of the particle with the medium. It is a vector function of time and usually termed "the white noise"<sup>14</sup>. It is assumed that the expected value of this function is identically equal to zero and that its individual components are mutually independent with the autocorrelation functions, the latter being equal to the Dirac function. The dot between the symbols designates the scalar product.

If the flow of the fluid itself displays random (turbulent) behaviour the symbol  $\mathbf{v}$  shall be taken to be the expected value of the velocity of the fluid that with the effect of random fluctuations of this velocity is incorporated in the tensor  $\mathbf{G}$ .

Eq. (1) is called the stochastic differential equation and it is written in the Langevine form. As long as the effect of the stochastic, i.e. the last term is negligible the equation changes into a set of three ordinary differential equations for individual coordinates of the vector  $\mathbf{X}(t)$ . In spite of the obvious physical interpretation it has, however, a substantial drawback. It may be proven that the vector  $\xi(t)$  is not correctly defined in the sense that each of its components in any time instant grows above all limits. In spite of this drawback, however, the stochastic differential equation in the Langevine form has been considered in many disciplines to be a useful tool. However, as noted by van Kampen<sup>12</sup>, the above notation does not have an unambiguous meaning; the properties of the vector  $\xi(t)$  are insufficient for a unique description of the process. This problem shall be discussed in the following paragraph.

#### *More Perfect Ways of Notation of Stochastic Differential Equations*

In order that we may remove the first of the drawbacks of the notation of Eq. (1), mentioned in the preceding paragraph, we shall introduce an integral of "white noise" as the, so called, Wiener process (see e.g. ref.<sup>2</sup>):

$$\mathbf{W}(t) = \int_0^t \xi(s) ds, \quad (2)$$

i.e. a random (vector) function of time with the probability density defined by

$$\begin{aligned} \frac{\partial^3}{\partial w_1 \partial w_2 \partial w_3} P\{W_1(t) < w_1, W_2(t) < w_2, W_3(t) < w_3\} = \\ = (2\pi t)^{-3/2} \exp[-(w_1^2 + w_2^2 + w_3^2)/2t]. \end{aligned} \quad (3)$$

From the given definition it follows that the Wiener process is a random function of time with the normal distribution, zero expected value and dispersion equal to the time that elapsed from the onset of the process. It is further apparent that in the initial time instant this process has a zero value. For the above stated reasons here we have directly defined the three-dimensional Wiener process; from the definition it also follows that the components of the process  $\mathbf{W}(t)$  are mutually independent.

By means of the Wiener process Ito has defined the stochastic integral (see e.g. ref.<sup>2</sup>) written in the unidimensional form as a limit of the sum

$$\int_{t_a}^{t_b} K(t) dW(t) = \lim_{\varrho \rightarrow 0} \sum_{k=0}^{n-1} K(t_k) [W(t_{k+1}) - W(t_k)], \quad (4)$$

where  $t_a = t_0 < t_1 \dots < t_n = t_b$ ,  $\varrho = \max(t_{k+1} - t_k)$ .

$K(t)$  is a general random function of time with certain properties; it must not firstly depend on the difference  $W(t_{k+1}) - W(t_k)$ . This requirement, however, is fully satisfied for a broad class of functions as with respect to the normal distribution of the Wiener process even the Wiener process  $W(t_k)$  itself, considered in the initial time instant of the time subinterval  $t_{k+1} - t_k$ , is independent of the increment  $W(t_{k+1}) - W(t_k)$ . First of all this property enabled the development of an extensive mathematical apparatus for the description of a large class of random processes (see e.g. refs<sup>2,13</sup>). It may be, for instance, shown that

$$\int_{t_a}^{t_b} K(t) dW^2(t) = \int_{t_a}^{t_b} K(t) dt; \quad \int_{t_a}^{t_b} K(t) dW(t) dt = 0; \quad (5)$$

$$\int_{t_a}^{t_b} K(t) dW_1(t) dW_2(t) = 0,$$

where  $W_1$  and  $W_2$  are mutually independent processes.

The definition of the stochastic integral (4) permits first of all correct notation of the stochastic differential equation (1) in the form

$$d\mathbf{X}(t) = \mathbf{v}^I(\mathbf{X}(t), t) dt + \mathbf{G}^I(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) \quad (6)$$

which has to be understood in the sense of the integral notation

$$\mathbf{X}(t_b) - \mathbf{X}(t_a) = \int_{t_a}^{t_b} \mathbf{v}^I(\mathbf{X}(t), t) dt + \int_{t_a}^{t_b} \mathbf{G}^I(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) \quad (7)$$

for all  $t_b$  and  $t_a$  satisfying the relation  $0 \leq t_a < t_b \leq T < \infty$ .

The first integral is the usual integral in the sense of the Riemann definition (for individual components of the vector  $\mathbf{v}^I$ ), the second is a stochastic integral in the sense of Ito calculus:

$$\int_{t_a}^{t_b} \mathbf{G}^I(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) = \lim_{\varrho \rightarrow 0} \sum_{k=0}^{n-1} \mathbf{G}^I(\mathbf{X}(t_k), t_k) \cdot [W(t_{k+1}) - W(t_k)]. \quad (8)$$

The coefficients in Eq. (6) have been distinguished by means of superscripts from the coefficients of Eq. (1); as it will be shown there need not be unique relationship between them. There exists namely another definition of the stochastic integral (4) or (8) proposed by Stratonovich, which shall be written directly in the three-dimen-

sional form

$$(S) \int_{t_a}^{t_b} \mathbf{G}^S(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) = \lim_{\rho \rightarrow 0} \sum_{k=0}^{n-1} \mathbf{G}^S((\mathbf{X}(t_{k+1}) + \mathbf{X}(t_k))/2, (t_{k+1} + t_k)/2) \cdot [\mathbf{W}(t_{k+1}) - \mathbf{W}(t_k)]. \quad (9)$$

In this definition the value of the function  $\mathbf{G}^S$  is assigned to the mean of the time subinterval  $t_{k+1} - t_k$  and it is thus dependent on the increment  $\Delta\mathbf{W}_k \equiv \mathbf{W}(t_{k+1}) - \mathbf{W}(t_k)$ . The integral (9), however, may be defined by means of the Ito integral (8) using a procedure<sup>13</sup> which shall be here introduced in a simpler manner. First we put  $\Delta\mathbf{X}_k \equiv \mathbf{X}(t_{k+1}) - \mathbf{X}(t_k)$  and  $\Delta t_k \equiv t_{k+1} - t_k$ , write the Taylor expansion of the tensor function  $\mathbf{G}^S$  with respect to these differences while neglecting all higher order differences excepting the first order differences:

$$\begin{aligned} \mathbf{G}^S((\mathbf{X}(t_{k+1}) + \mathbf{X}(t_k))/2, (t_{k+1} + t_k)/2) &= \mathbf{G}^S(\mathbf{X}(t_k) + \Delta\mathbf{X}_k/2, t_k + \Delta t_k/2) \\ &\approx \mathbf{G}^S(\mathbf{X}(t_k), t_k) + \frac{1}{2}[\Delta\mathbf{X}_k \cdot \nabla] \mathbf{G}^S(\mathbf{x}, t_k)|_{\mathbf{x}=\mathbf{X}(t_k)} + \\ &\quad + \frac{\Delta t_k}{2} \frac{\partial}{\partial t} \mathbf{G}^S(\mathbf{X}(t_k), t)|_{t=t_k} + \dots \end{aligned} \quad (10)$$

where

$$\nabla \equiv \mathbf{e}_1 \frac{\partial}{\partial x_1} + \mathbf{e}_2 \frac{\partial}{\partial x_2} + \mathbf{e}_3 \frac{\partial}{\partial x_3}$$

designates the differential operator in the three-dimensional Euclid space and the brackets the priority of mathematical operations. In the following text we shall always consider that the operator  $\nabla$  relates to all terms of the product written to the right of this symbol.

For the difference  $\Delta\mathbf{X}_k$  we shall substitute from the stochastic differential equation (6) considering the integral form (7)

$$\begin{aligned} \mathbf{G}_k^S + \frac{1}{2}[\Delta\mathbf{X}_k \cdot \nabla] \mathbf{G}_k^S + \frac{\Delta t_k}{2} \frac{\partial}{\partial t} \mathbf{G}_k^S &\approx \mathbf{G}_k^S + \frac{1}{2}[\mathbf{v}_k^I \Delta t_k \cdot \nabla] \mathbf{G}_k^S + \\ &+ \frac{1}{2}[\mathbf{G}_k^I \cdot \Delta\mathbf{W}_k \cdot \nabla] \mathbf{G}_k^S + \frac{\Delta t_k}{2} \frac{\partial}{\partial t} \mathbf{G}_k^S + \dots \end{aligned} \quad (11)$$

All functions in the last equation are assigned to the beginning of the time subinterval  $\Delta t_k$ , in the abbreviated notation we put  $\mathbf{G}_k^S \equiv \mathbf{G}^S(\mathbf{X}(t_k), t_k)$ ; further it hold  $\mathbf{v}_k^I \equiv \mathbf{v}^I(\mathbf{X}(t_k), t_k)$  and  $\mathbf{G}_k^I \equiv \mathbf{G}^I(\mathbf{X}(t_k), t_k)$ . From the expressions (10) and (11) substituted into Eq. (9) it is apparent that with respect to the second of Eqs (5) the terms in product with  $\Delta t_k$  are equal to zero. Therefore we shall obtain again in the ab-

breviated notation

$$(S) \int_{t_a}^{t_b} \mathbf{G}^S \cdot d\mathbf{W} = \int_{t_a}^{t_b} \mathbf{G}^S \cdot d\mathbf{W} + \frac{1}{2} \int_{t_a}^{t_b} [\mathbf{G}^I \cdot d\mathbf{W} \cdot \nabla] \mathbf{G}^S \cdot d\mathbf{W}. \quad (12)$$

The two integrals on the right hand side are the Ito integrals defined by Eq. (8).

In the last term of the expression behind the integral we shall perform first a few operations of the scalar multiplication. The symbol  $\mathbf{G}^+$  shall designate the tensor whose matrix is a transpose matrix with respect to the tensor  $\mathbf{G}$ , i.e.  $\{G_{pq}\} = \{G_{qp}^+\}$  for each element of the matrix. Thus we obtain

$$\begin{aligned} \int_{t_a}^{t_b} [\mathbf{G}^I \cdot d\mathbf{W} \cdot \nabla] \mathbf{G}^S \cdot d\mathbf{W} &= \int_{t_a}^{t_b} [d\mathbf{W} \cdot \mathbf{G}^{+I} \cdot \nabla] d\mathbf{W} \cdot \mathbf{G}^{+S} = \\ &= \int_{t_a}^{t_b} [\mathbf{G}^{+I} \cdot \nabla \cdot d\mathbf{W}] d\mathbf{W} \cdot \mathbf{G}^{+S} = \int_{t_a}^{t_b} [\mathbf{G}^{+I} \cdot \nabla \cdot I] dt \cdot \mathbf{G}^{+S} = \\ &= \int_{t_a}^{t_b} [\mathbf{G}^{+I} \cdot \nabla] \cdot \mathbf{G}^{+S} dt \end{aligned} \quad (13)$$

The expression  $d\mathbf{W} d\mathbf{W}$  in the middle of these equations is a dyadic product which may be described by the matrix whose elements are equal  $\{W_p(t) W_q(t)\}$ ,  $p, q = 1, 2, 3$ . From the third of the expressions (5) it is apparent that for  $p \neq q$  the corresponding integrals vanish; the case  $p = q$  is determined by the first of the relations (5). Finally we shall consider that the functions  $\mathbf{G}^I$  and  $\mathbf{G}^S$  are identical, i.e. that for each element of the matrix we have  $\{G_{pq}^I(\mathbf{X}(t), t)\} \equiv \{G_{pq}^S(\mathbf{X}(t), t)\}$ , so that we obtain the relation for the Stratonovich integral in dependence on the Ito integral

$$(S) \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) = \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) + (1/2) \int_{t_a}^{t_b} \mathbf{J}(t) dt, \quad (14)$$

where the superscripts are omitted and where the vector

$$\mathbf{J}(t) \equiv [\mathbf{G}^+(\mathbf{X}(t), t) \cdot \nabla] \cdot \mathbf{G}(\mathbf{X}(t), t) \quad (15)$$

shall be termed the "semidiffusion" flux. By means of the integral, defined in such a way, one can write the Stratonovich differential equation in the form

$$d\mathbf{X}(t) = \mathbf{v}^S(\mathbf{X}(t), t) dt + \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t). \quad (S) \quad (16)$$

As long as  $\mathbf{X}(t)$  is to be the same solution of both equations we must have that

$$\mathbf{v}^S(\mathbf{X}(t), t) = \mathbf{v}^I(\mathbf{X}(t), t) - (1/2)\mathbf{J}(t). \quad (17)$$

The presented results may be derived mathematically quite rigorously by introducing the so-called Q-multiplication in abstract spaces<sup>15</sup>.

In the very same way we shall now introduce the third definition of the stochastic

integral by assigning the subintegral function to the end of the time subintervals  $t_{k+1} - t_k$ :

$$(T) \int_{t_a}^{t_b} \mathbf{G}^T(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) = \lim_{q \rightarrow 0} \sum_{k=0}^{n-1} \mathbf{G}^T(\mathbf{X}(t_{k+1}), t_{k+1}) \cdot [\mathbf{W}(t_{k+1}) - \mathbf{W}(t_k)]. \quad (18)$$

The integral defined in such a way shall be termed the transport stochastic integral for – as we shall further show – the stochastic differential equation in which one can use this integral is adequate to the differential equations applied in chemical engineering for the description of mass and heat transfer.

Further we shall use the analogous procedure as in the case of the Stratonovich integral. Instead of Eq. (10) we shall obtain

$$\begin{aligned} \mathbf{G}^T(\mathbf{X}(t_{k+1}), t_{k+1}) &= \mathbf{G}^T(\mathbf{X}(t_k) + \Delta\mathbf{X}_k, t_k + \Delta t_k) \approx \mathbf{G}^T(\mathbf{X}(t_k), t_k) + \\ &+ [\Delta\mathbf{X}_k \cdot \nabla] \mathbf{G}^T(\mathbf{x}, t_k)|_{\mathbf{x}=\mathbf{X}(t_k)} + \Delta t_k \frac{\partial}{\partial t} \mathbf{G}^T(\mathbf{X}(t_k), t)|_{t=t_k} + \dots \end{aligned} \quad (19)$$

It is apparent that the expressions in Eqs (11) through (17) shall be analogous; only the coefficient 1/2 disappears in the corresponding terms. We may thus write, instead of Eq. (14), the relation between the Ito and the transport integral and considering the same equation also the relation between the Stratonovich integral

$$\begin{aligned} (T) \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) &= \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) + \int_{t_a}^{t_b} \mathbf{J}(t) dt = \\ (S) \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) &+ \int_{t_a}^{t_b} \mathbf{J}(t) dt/2. \end{aligned} \quad (20)$$

The stochastic differential equation on the basis of the transport integral has an analogous form (16)

$$d\mathbf{X}(t) = \mathbf{v}^T(\mathbf{X}(t), t) dt + \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t), \quad (T) \quad (21)$$

where

$$\mathbf{v}^T(\mathbf{X}(t), t) = \mathbf{v}^I(\mathbf{X}(t), t) - \mathbf{J}(t) = \mathbf{v}^S(\mathbf{X}(t), t) - \mathbf{J}(t)/2. \quad (22)$$

By means of the Wiener process it could be clearly possible to define the whole family of stochastic integrals<sup>13</sup> and corresponding stochastic differential equations in dependence on the choice of the time instant  $\Delta t_k$ , i.e.

$$\begin{aligned} (\alpha) \int_{t_a}^{t_b} \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) &= \lim_{q \rightarrow 0} \sum_{k=0}^{n-1} \mathbf{G}(\mathbf{X}(t_k) + \alpha \Delta\mathbf{X}_k, t_k + \alpha \Delta t_k) \cdot \\ &\cdot [\mathbf{W}(t_{k+1}) - \mathbf{W}(t_k)]. \quad [0 \leq \alpha \leq 1] \end{aligned} \quad (23)$$

The integrals for  $\alpha = 0$  and  $\alpha = 1/2$  (the Ito and the Stratonovich integral) play an important role in the theory of random processes as well as in practical applications. In the following paragraph we shall attempt to show also the possible applications of the transport integral, i.e. the case when  $\alpha = 1$ .

*The Relationships between Stochastic Differential Equations  
and the Kolmogorov Equations*

We shall consider again a random motion of a distinguishable particle (molecule of the component) in the flowing fluid. The relationship (13.I)\* in the preceding communication<sup>1</sup> defined the transitive probability density  $f(\mathbf{x}; t|\mathbf{y}; \tau)$  that the particle with the probability  $f dV$  shall appear in an infinitesimally small volume  $dV = dx_1 dx_2 dx_3$  on condition that at some previous time instant  $\tau < t$  the particle certainly was at the point determined by the position vector  $\mathbf{y}$ .

Let us assume further that the Ito stochastic differential equation (6) describes this motion with sufficient accuracy so that we may write

$$\mathbf{X}(t) = \mathbf{y} + \int_{\tau}^t \mathbf{v}^I(\mathbf{X}(s), s) ds + \int_{\tau}^t \mathbf{G}(\mathbf{X}(s), s) \cdot d\mathbf{W}(s) \quad (24)$$

In the literature (see e.g. ref.<sup>2</sup>) it is proven that for the transitive probability density of this process  $f = f(\mathbf{x}; t|\mathbf{y}; \tau)$  one can derive the Kolmogorov forward diffusion equation (parabolic differential equation) which we shall write in the following form

$$\partial f / \partial t + \nabla \cdot [\mathbf{v}^I(\mathbf{x}, t) f] - (1/2) \nabla \cdot [\nabla \cdot [\mathbf{G}(\mathbf{x}, t) \cdot \mathbf{G}^+(\mathbf{x}, t)] f] = 0. \quad (25)$$

(For the meaning of the brackets and the function of the differential operator see the note past Eq. (10)). The written equation is identical with (18.I) while it holds

$$\mathbf{v}^I(\mathbf{x}, t) \equiv \mathbf{a}(\mathbf{x}, t); \quad \mathbf{G}(\mathbf{x}, t) \cdot \mathbf{G}^+(\mathbf{x}, t) \equiv \mathbf{B}(\mathbf{x}, t). \quad (26)$$

For individual elements of the matrix we may write

$$\{B_{pq}(\mathbf{x}, t)\} = \left\{ \sum_{r=1}^3 G_{pr}(\mathbf{x}, t) G_{qr}(\mathbf{x}, t) \right\} \quad ,$$

and Eq. (25) may be written in the component form

$$\frac{\partial f}{\partial t} + \sum_{p=1}^3 \frac{\partial}{\partial x_p} [v_p^I(\mathbf{x}, t) f] - (1/2) \sum_{p,q=1}^3 \frac{\partial^2}{\partial x_p \partial x_q} [B_{pq}(\mathbf{x}, t) f] = 0. \quad (25a)$$

\* The relationship presented in the preceding communication<sup>1</sup> shall be here referred to by the form (K. I) where K is the number of the equation in the first communication.



In case that the Stratonovich stochastic differential equation (16) is regarded as more accurate we shall use again the previous procedure with that in the Ito equation (24) we substitute for  $\mathbf{v}^I$  from Eq. (17):

$$\mathbf{X}(t) = \mathbf{y} + \int_{\tau}^t \mathbf{v}^S(\mathbf{X}(s), s) ds + (1/2) \int_{\tau}^t J(s) ds + \int_{\tau}^t \mathbf{G}(\mathbf{X}(s), s) \cdot d\mathbf{W}(s). \quad (27)$$

Now we are in the position to write down the analog of Eq. (25) by performing the inner differentiation of the product in the last term in such a way that the operator shall first operate on the function  $\mathbf{G}^+$  and then on the product  $\mathbf{G}f$ . Thus we obtain, after some modifications

$$\partial f / \partial t + \nabla f \cdot [\mathbf{v}^S + (1/2) J] - (1/2) \nabla \cdot [Jf + \mathbf{G} \cdot [\nabla \cdot \mathbf{G}] f] = 0. \quad (28)$$

After multiplication the terms containing the semidiffusion flux  $J$  clearly cancel out so that we finally obtain the Kolmogorov equation corresponding to the Stratonovich equation

$$\partial f / \partial t + \nabla f \cdot \mathbf{v}^S - (1/2) \nabla \cdot [\mathbf{G} \cdot [\nabla \cdot \mathbf{G}] f] = 0, \quad (29)$$

or in the component notation

$$\frac{\partial f}{\partial t} + \sum_{p=1}^3 \frac{\partial}{\partial x_p} [v_p^S(\mathbf{x}, t) f] - (1/2) \sum_{p,q,r=1}^3 \frac{\partial}{\partial x_p} \left[ G_{pr}(\mathbf{x}, t) \frac{\partial}{\partial x_q} [G_{qr}(\mathbf{x}, t) f] \right] = 0. \quad (29a)$$

Finally we can write down analogously, provided that the transport stochastic equation (21) describes best the process considered, Eq. (24) after substituting here from Eq. (22)

$$\mathbf{X}(t) = \mathbf{y} + \int_{\tau}^t \mathbf{v}^T(\mathbf{X}(s), s) ds + \int_{\tau}^t J(s) ds + \int_{\tau}^t \mathbf{G}(\mathbf{X}(s), s) \cdot d\mathbf{W}(s). \quad (30)$$

Adequate Kolmogorov equation is similar to Eq. (28) in which we still differentiate the product  $\mathbf{G}f$  in the last term and at the same time subtract the terms containing the semidiffusion flux  $J$ ; thus we obtain

$$\partial f / \partial t + \nabla f \cdot \mathbf{v}^T + (1/2) \nabla f \cdot [J - \mathbf{G} \cdot [\nabla \cdot \mathbf{G}]] - (1/2) \nabla \cdot [[\mathbf{G} \cdot \mathbf{G}^+] \cdot \nabla f] = 0, \quad (31)$$

or in the component notation

$$\begin{aligned} \frac{\partial f}{\partial t} + \sum_{p=1}^3 \frac{\partial}{\partial x_p} v_p^T f + (1/2) \sum_{p=1}^3 \frac{\partial}{\partial x_p} f \left[ \sum_{q,r=1}^3 \left[ \frac{\partial G_{pr}}{\partial x_q} G_{qr} - G_{pr} \frac{\partial G_{qr}}{\partial x_q} \right] \right] - \\ - (1/2) \sum_{p,q=1}^3 \frac{\partial}{\partial x_p} \left[ B_{pq} \frac{\partial f}{\partial x_q} \right] = 0. \end{aligned} \quad (31a)$$

In the last term we took into consideration the second from the definition relationship (26). Clearly, as follows from the previous communication<sup>1</sup> in case that the third term should be equal to zero the relationship (31) would be, as far as the position of the differential operators is concerned, identical with the transport equations (11.I) and (12.I) usually used in chemical engineering. The following relationship would then have to hold for all  $p, q, r = 1, 2, 3$

$$\frac{\partial G_{pr}}{\partial x_q} G_{qr} = G_{pr} \frac{\partial G_{qr}}{\partial x_q}. \quad (32)$$

It can be easily shown that in this case the elements of the matrix of the tensor  $\mathbf{G}$  would have to have the following form

$$\{G_{pq}(\mathbf{x}, t)\} = \{c_{pq}g_q(\mathbf{x}, t)\}, \quad (33)$$

where  $c_{pq}$  are constants (or functions of time only) and  $g_q$  are generally scalar functions of spatial coordinates and time which are the same in individual columns of the matrix. It is further apparent that the condition is immediately fulfilled as long as  $\mathbf{G}$  is independent of spatial coordinates. Some coefficients, however, may equal zero so that Eq. (32) is, for instance, fulfilled for each diagonal matrix.

The elements of the matrix of the tensor  $\mathbf{B}$  are in the general case a linear combination of the functions  $g$ , i.e.

$$\{B_{pq}(\mathbf{x}, t)\} = \left\{ \sum_{r=1}^3 c_{pr}c_{qr}g_r^2(\mathbf{x}, t) \right\}; \quad (34)$$

the condition (32) is clearly met in the important case when the coefficient  $\mathbf{B}$  is a scalar

$$\mathbf{B}(\mathbf{x}, t) = IB(\mathbf{x}, t), \quad (35)$$

i.e. in the case of inhomogeneous isotropic diffusion.

For a sufficiently broad class of processes Eq. (31) may thus be written in the following form

$$\partial f / \partial t + \nabla[f \cdot \mathbf{v}^T] - (1/2) \nabla \cdot [\mathbf{B} \cdot \nabla f] = 0 \quad (36)$$

with simultaneous validity of Eq. (32), or, if Eqs (33) and (34) or (35) hold. After integration indicated in Eq. (19.I) this equation holds also for the unconditional probability  $p(\mathbf{x}; t)$  which is, in view of Eqs (20.I) or (24.I) under certain simplifying assumptions, proportional to the concentration of the component or the temperature of the fluid. The expression  $\mathbf{B}(\mathbf{x}, t)/2$  may then be regarded, in view of Eqs (23.I) or (25.I), as the diffusion tensor or tensor of thermal diffusivity.

## DISCUSSION

In the preceding paragraphs we have presented three ways of notation of the stochastic integral from which there follow three ways of notation of the stochastic differential equations and the corresponding Kolmogorov equations. From the mathematical standpoint all three relations are equivalent as their results are in all cases identical solutions (in the probabilistic sense), i.e. functions  $\mathbf{X}(t)$ , or the transitive probability density  $f(\mathbf{x}; t|\mathbf{y}; \tau)$  for this function are identical.

Problems with utilization of relations arise in physical applications and are due to — as noted by Seinfeld and Lapidus<sup>4</sup> — the pathological character of the Wiener process which has no derivatives and as such is unrealizable. For this reason some authors avoid using this mathematical apparatus<sup>16</sup> in describing random processes in chemical engineering. Of course, it is obvious that these problems appear only in case that the stochastic term (tensor  $\mathbf{G}$ ) is an explicit function of the position vector  $\mathbf{X}(t)$ .

The procedure in setting up a chemical engineering or other model is usually such that one sets up first the description of its deterministic components. On this one superimposes (in case of diffusion processes considered in the probabilistic sense, i.e. random processes for which conditions (15–17.I) hold) as a randomising factor the term containing the white noise function or the differential of the Wiener process without deeper analysis of this term. Such a procedure then leads to different results as a deeper analysis of the stochastic term is usually practically impossible.

In our case considered, i.e. in the study of the kinematics of the random motion of a particle in the flowing fluid, which may lead to notation of the transport equations, we thus take the symbols  $\mathbf{v}$  in Eqs (1), (6), (15), and (21) as the velocity of the fluid, i.e. as identical functions of spatial coordinates and time.

Such a procedure has been used also in the previous communication<sup>1</sup>; we have presented also a review of the cases leading to identical solutions of the Kolmogorov equations and the transport equations (i.e. differential mass and enthalpy balances). The above review is clearly valid also for the Kolmogorov equation (29) corresponding to the Stratonovich stochastic differential equation. In this connection one has to remind that van Kampen's remark (see ref.<sup>12</sup>, p. 291) that this relationship describes inhomogeneous diffusion is erroneous as follows from comparison of Eqs (3.1) and (3.2) in the cited work and unidimensional variants of Eqs (25), (29), and (31) of this paper.

In the previous communication<sup>1</sup> we have also stated that apparently it cannot be unambiguously decided which of Eqs (25) or (31) — generally the transport equations — is “better” and that one has always to consider, or experimentally verify, the concrete situation. The same is true also with the use of Eq. (29). It seems that in the majority of chemical engineering applications one should prefer Eq. (31) as this is the only equation leading to the uniform distribution of the probability

density in the steady state and bounded space with reflecting boundaries (see e.g. ref.<sup>17</sup>). Physically this situation corresponds to the uniform distribution of concentration of the mass component or temperature in the bounded part of the space under the steady state. It has to be noted though that in case that the condition (32) is invalid or in cases when the Kolmogorov equation in the form (25) cannot be transformed to the form (36) (see ref.<sup>1</sup>), the transport equations cannot be derived on the basis of the concept of the stochastic processes developed here.

In connection with the development of direct modelling of stochastic differential equations on the computers one has to point at yet another fact. It seems that from the viewpoint of a numeric experiment one should prefer the Stratonovich way of notation of the stochastic differential equations<sup>10,11,18</sup> which may be due to the "symmetric" definition of the corresponding integral. (In the original work<sup>14</sup> Stratonovich proved, in fact without using this definition, the equivalency of Eqs (1) and (16)).

In this case it would be necessary to use in the modelling of chemical engineering processes leading to the uniform distribution the relationship transforming Eq. (21) into (16), i.e. to model the process with the aid of the following relationship

$$d\mathbf{X}(t) = (\mathbf{v}^T(\mathbf{X}(t), t) + \mathbf{J}(t)/2) dt + \mathbf{G}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t) \quad (\text{S}), \quad (37)$$

where  $\mathbf{J}$  is determined by Eq. (15). The stochastic term in this equation is determined by the Stratonovich integral; the function  $\mathbf{G}$  must, of course, satisfy condition (32).

In conclusion we shall introduce the approach which, although substantially more complicated, is free of the analysed difficulties. The problems that here arise are from the physical standpoint caused, among others, by the fact that the kinematic model described in Eq. (1), or by analogous equations, is considerably inaccurate. We shall therefore introduce the "dynamic" model enabling us to consider generally also the forces acting on the particle

$$m d\mathbf{V}(t)/dt = -\beta[\mathbf{V}(t) - \mathbf{v}(\mathbf{X}(t), t)] + m\mathbf{h}(\mathbf{X}(t)) + \mathbf{N}(\mathbf{X}(t), t) \cdot \xi(t), \quad (38)$$

$$d\mathbf{X}(t)/dt = \mathbf{V}(t). \quad (39)$$

By means of this Langevine equation one can characterise not only the position but also the velocity  $\mathbf{V}$  of the particle. Eq. (38) is a way of writing the second Newton law; it is assumed that the particle is under the action of the friction forces (directly proportional – with the constant coefficient of proportionality  $\beta$  – to the difference between the velocity of the particle and the fluid), the external conservative force, proportional to its mass,  $m$ , and further the resulting force of random character, expressed by the last term.

It can be easily proven that as long as the coefficient  $\mathbf{N}$  in the stochastic term is not an explicit function of the velocity, all stochastic integrals, defined in the previous paragraphs, are identical. The increment  $\Delta \mathbf{X}_k$ , for instance in Eq. (10), is in this case on the basis of Eqs (39) proportional only to the time difference  $\mathbf{V}(t_k) \Delta t_k$ . Thus after substituting into the Ito stochastic integral this term vanishes with respect to the second one of Eqs (5).

If we put, in addition, that  $\gamma \equiv \beta/m$ ;  $\mathbf{H}(\mathbf{X}(t), t) \equiv \mathbf{N}(\mathbf{X}(t), t)/m$  we can write Eq. (38) in a unique form

$$d\mathbf{V}(t) = -\gamma[\mathbf{V}(t) - \mathbf{v}(\mathbf{X}(t), t)] dt + \mathbf{h}(\mathbf{X}(t)) dt + \mathbf{H}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t), \quad (40)$$

to which corresponds, in view of Eq. (39), also the only Kolmogorov equation,

$$\begin{aligned} \partial f_2 / \partial t + \mathbf{u} \cdot \nabla f_2 - \gamma \nabla_{\mathbf{u}} \cdot (\mathbf{u} f_2) + (\gamma \mathbf{v}(\mathbf{x}, t) + \mathbf{h}(\mathbf{x})) \cdot \nabla_{\mathbf{u}} f_2 - \\ - (1/2) \mathbf{H}(\mathbf{x}, t) \cdot \mathbf{H}^+(\mathbf{x}, t) : \nabla_{\mathbf{u}}^2 f_2 = 0, \end{aligned} \quad (41)$$

where  $f_2 = f_2(\mathbf{x}, \mathbf{u}; t | \mathbf{y}, \mathbf{u}_0; \tau)$  denotes the six-dimensional transitive probability density characterising both the velocity as well as the position of the particle.  $\nabla_{\mathbf{u}}$  designates the differential operator in the velocity domain and  $\nabla_{\mathbf{u}}^2$  the dyadic product of these operators. The colon is a double scalar product.

This equation, although substantially more complicated, permits one to express also the effect of the intensity of external forces,  $\mathbf{h}$ , on this particle. Let us consider further that the mass of the particle is very small so that the ratio of the friction coefficient and its mass, equally as the tensor  $\mathbf{H}$ , assume disproportionately larger values in comparison with this intensity, which shall be therefore neglected. Further we shall substitute from Eq. (39) into (40) and obtain

$$d\mathbf{V}(t) + \gamma d\mathbf{X}(t) = \gamma \mathbf{v}(\mathbf{X}(t), t) dt + \mathbf{H}(\mathbf{X}(t), t) \cdot d\mathbf{W}(t). \quad (42)$$

For large values of  $\gamma$  it may be assumed that the velocity of the particle relaxes to the steady state value (and hence  $d\mathbf{V} \rightarrow 0$ ) in contrast to the "slow" variations of  $d\mathbf{X}$  and therefore for longer times elapsed from the onset of the process we neglect the velocity fluctuations of the particle with respect to the fluctuations of its position (This procedure is termed adiabatic elimination<sup>13</sup>). It is apparent that with this procedure Eq. (42) does not differ from the earlier written stochastic differential equations (we have  $\mathbf{G} \equiv \mathbf{H}/\gamma$ ), where, however, problems arise with the definition of the stochastic integral.

Eqs (39)–(41) for the case  $\mathbf{h}(\mathbf{x}) = \mathbf{0}$  in this sense may be regarded as unambiguous description of the motion of the particle in the flowing fluid whose velocity is in this case always determined by the symbols  $\mathbf{v}(\mathbf{x}, t)$ . The transitive probability density

$f(\mathbf{x}; t|\mathbf{y}; \tau)$ , determined by Eq. (13.1) may be obtained by integration of the function  $f_2$  in the velocity domain and after introducing the assumption that the particle had at the initial time instant  $\tau$  zero velocity:

$$f(\mathbf{x}; t|\mathbf{y}; \tau) = \int f_2(\mathbf{x}, \mathbf{u}; t|\mathbf{y}, \mathbf{0}; \tau) d\mathbf{u}. \quad (43)$$

In the Appendix of this paper we have presented a simple example illustrating the outlined procedure. Its results, however, cannot be physically interpreted, yet as an advantage remains that all solutions may be written in quadratures.

## CONCLUSIONS

From the considerations presented in this paper one can draw the following conclusions:

1. A third definition of the stochastic integral, termed the transport integral, (Eq. (18)) has been proposed on the basis of the relationship between the Ito and the Stratonovich definition. Relationships between these three types have been written down (Eq. (20)). The transport integral enables us to write the stochastic differential equation (Eq. (21)) and the corresponding Kolmogorov forward equation (Eq. (31)) which is, under certain confining conditions (Eq. (32)), formally identical with the differential equations describing the transport of mass and heat.

2. It was shown that the different results provided by these three types of the stochastic differential equations (the Ito, the Stratonovich and the transport equation) and the adequate Kolmogorov equations, in case that the stochastic term is a function of the spatial coordinates, are due to the different interpretation of individual terms in these equations.

3. An equation has been proposed, Eq. (37), permitting us to model the transport phenomena usually considered in chemical engineering by means of stochastic differential equation while maintaining the validity of the conditions (32).

4. A "dynamic" model has been proposed, explicitly incorporating also the rate of the process and a corresponding equation has been written down, Eq. (40), which though substantially more complex, enables a unified interpretation of individual terms and hence to obtain always the same results independently of the definition of the stochastic integral. A simple example has been presented of such a procedure (see the Appendix).

## APPENDIX

### *Various Forms of the Diffusion Equation and their Solutions*

We shall first write down a unidimensional analog of the stochastic differential equation (39) and (40) in the form

$$dV(t) = -V(t) dt + (1 + p(X(t)))^{1/2} dW(t); \quad dX(t) = V(t) dt, \quad (D.1)$$

where  $p(x)$  is a so far undefined function and  $v(x, t) \equiv 0$ .

The Kolmogorov equation corresponding to this equation is

$$\partial f_2 / \partial t + u \partial f_2 / \partial x - \partial(u f_2) / \partial u - (1/2)(1 + p(x)) \partial^2 f_2 / \partial u^2 = 0. \quad (D.2)$$

Using the method of adiabatic elimination we shall obtain a simpler stochastic differential equation

$$dX(t) = (1 + p(X(t)))^{1/2} dW(t), \quad (D.3)$$

with the three corresponding Kolmogorov equations as long as we put

$$v^I = v^S = v^T = v(x, t) = 0.$$

The Ito interpretation

$$\partial f / \partial t - (1/2) \partial^2 ((1 + p(x)) f) / \partial x^2 = 0; \quad (D.4)$$

the Stratonovich interpretation

$$\frac{\partial f}{\partial t} - (1/2) \frac{\partial}{\partial x} ((1 + p(x))^{1/2} \frac{\partial}{\partial x} (1 + p(x))^{1/2} f) = 0; \quad (D.5)$$

the "transport" interpretation

$$\partial f / \partial t - (1/2) \frac{\partial}{\partial x} \left( (1 + p(x)) \frac{\partial f}{\partial x} \right) = 0. \quad (D.6)$$

It is apparent that the coefficient  $1 + p(x)$  satisfies the condition (32).

We shall now deal with only the steady state solution of Eqs (D.2), (D.4-6). Eq. (D.2) shall be rewritten into the following form

$$u \partial f_2 / \partial x = \partial(u f_2) / \partial u + (1/2)(1 + p(x)) \partial^2 f_2 / \partial u^2. \quad (D.7)$$

Solution of this equation shall be sought as the normal distribution of the variable  $u$

$$f_2 = f_2(x, u) = f(x) \exp(-(u - q(x))^2 / 2) / (2\pi)^{1/2}. \quad (D.8)$$

Having performed the differentiations and substitution into Eq. (D.7) the coefficients of individual power of  $u$  must be equal: After some modification we shall obtain the following relationship:

The coefficients of  $u^2$ :  $dq/dx = (p - 1)/2$ ;

$$u^1: d \ln f / dx = -q(p + 1)/2 ; \quad (D.9)$$

$$u^0: q^2 = (p - 1)/(p + 1) .$$

From the first and the third relation (D.9) we shall eliminate  $q$ ; after some modifications we obtain

$$2 dp/dx = \pm(p^2 - 1)^{3/2} , \quad (D.10)$$

and further we chose only the negative root. It is apparent that the choice of the form of the solution (D.8) determines the form of the function  $p(x)$ . After integration of Eq. (10) and some modifications we obtain, putting the integration constant equal to zero:

$$p(x) = x/(x^2 - 4)^{1/2} . \quad (D.11)$$

Further we shall inspect only the solution in region  $x \geq 2$ . From Eq. (D.8) hold the following relations

$$\int_{-\infty}^{+\infty} f_2 du = f(x) ; \quad \int_{-\infty}^{+\infty} u f_2 du = q(x) f(x) ; \quad (D.12)$$

the first of these relations is a simplification of Eq. (43). Integrating in this way the whole equation (D.7) we obtain

$$d(f(x) q(x))/dx = 0 ; \quad f(x) q(x) = K = \text{const} , \quad (D.13)$$

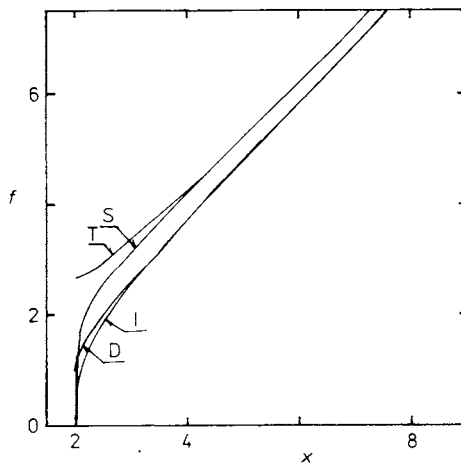


FIG. 1

Graphic illustration of the solutions of various forms of the diffusion equations. D "dynamic" model (Eqs (D.2), (D.8)); I Ito interpretation (Eqs (D.4), (D.19)); S Stratonovich interpretation (Eqs (D.5), (D.20)); T "transport" interpretation (Eqs (D.6), (D.21))



as all terms on the right hand side of Eq (D 7) after integration vanish. From the second of equations (D.9) we thus obtain the following relation for  $f(x)$

$$df/dx = -K(p + 1)/2. \quad (D.14)$$

The steady state solutions of Eqs (D 4–6) shall be designated  $f^I$ ,  $f^S$ ,  $f^T$  and after the first integration we write

$$d((p + 1)f^I)/dx = -2K \quad (D.15)$$

$$d((p + 1)^{1/2}f^S)/dx = -2K(p + 1)^{-1/2} \quad (D.16)$$

$$df^T/dx = -2K(p + 1)^{-1}. \quad (D.17)$$

The constant in Eqs (D.14–17) has the same value; from the physical standpoint it represents the intensity of diffusion flow. We put  $K = -1$ ,  $s = (x^2 - 4)^{1/2}$ ;  $r = x - s$ . The solution of all mentioned relationships may be written in the form of quadratures:

$$f = (s + x)/2 \quad (D.18)$$

$$f^I = xsr/2 \quad (D.19)$$

$$f^S = [3(2sr)^{1/2} \arcsin(r/2) + s(4 + r^2/2)]/4 \quad (D.20)$$

$$f^T = (s^3 - x^3 + 12x)/6. \quad (D.21)$$

It may be easily proven that for large values of  $x$  all solutions are identical

$$f = f^I = f^S = f^T = x \quad [x \rightarrow \infty]. \quad (D.22)$$

In the proximity of the point  $x = 2$ , however, they differ little. If we consider  $f(x)$  as “correct” then, as may be apparent from Fig. 1, the nearest solution is the one according to Ito –  $f^I(x)$ .

#### LIST OF SYMBOLS

<b>a</b>	drift velocity, $m s^{-1}$
<b>B</b>	diffusion tensor, $m^2 s^{-1}$
<b>c</b>	constant coefficients of matrix of tensor <b>G</b> , $m s^{-1/2}$
<b>e</b>	unit vector
<b>f</b>	transitive probability density of particle position, $m^{-3}$
<b>f<sub>2</sub></b>	transitive probability density of particle position and velocity, $m^{-6} s^3$
<b>G</b>	stochastic tensor, $m s^{-1/2}$
<b>g</b>	scalar function in columns of matrix of tensor <b>G</b>
<b>H</b>	tensor of “intensity” of random forces, $m s^{-3/2}$

$h$	intensity of conservative forces, $m s^{-2}$
$I$	identity tensor
$J$	"semidiffusion" flux, $m s^{-1}$
$K$	random function of time independent of $\Delta W$
$m$	mass of particle, kg
$N$	tensor of random forces, $kg m s^{-3/2}$
$p$	unconditional probability density for particle position, $m^{-3}$
$t$	time, s
$u$	particle velocity — variable in distribution function, $m s^{-1}$
$V$	volume, $m^3$
$V$	particle velocity, $m s^{-1}$
$v$	fluid velocity, $m s^{-1}$
$W$	Wiener process (three-dimensional) — random function of time, $s^{1/2}$
$w$	variable in distribution function of Wiener process, $s^{1/2}$
$X$	particle position vector — random function of time, m
$x$	particle position vector — variable in distribution function, m
$y$	initial particle position, m
$\beta$	coefficient of friction, $kg s^{-1}$
$\gamma$	"intensity" of coefficient of friction, $s^{-1}$
$\xi$	"white noise" function, $s^{-1/2}$
$\tau$	initial time instant, s
$\nabla$	differential operator with respect to spatial coordinates, $m^{-1}$
$\nabla_u$	differential operator with respect to velocities, $m^{-1} s$
$\Delta$	difference (increment)

## Subscripts

$a$	related to beginning of integration time interval
$b$	related to end of integration time interval
$k$	related to beginning of $k$ -th time subinterval
$0$	related to initial velocity
$p$	index of matrix of tensor
$q$	index of matrix of tensor
$r$	index of matrix of tensor

## Superscripts

$I$	related to Ito interpretation
$S$	related to Stratonovich interpretation
$T$	related to "transport" interpretation
$G^+$	tensor whose matrix is transpose with respect to matrix of tensor $G$

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Translated by V. Staněk.