

## Effect of Chemical Reactions and Phase Transitions on Turbulent Transport of Particles and Gases

Tov Elperin, Nathan Kleorin, and Igor Rogachevskii

*The Pearlstone Center for Aeronautical Engineering Studies, Department of Mechanical Engineering, Ben-Gurion University of the Negev, P.O. Box 653, Beer-Sheva 84105, Israel*

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Strong modification of turbulent transport in fluid flows with chemical reactions or heterogeneous phase transitions is analyzed. It is shown that turbulent diffusion may be strongly depleted in chemically reacting flows or in flows with phase transitions. In addition, the existence of nondiffusive heat and mass fluxes of the components in the systems with chemical reactions or heterogeneous phase transitions is demonstrated. These fluxes can be interpreted as “turbulent” analogs of the known cross-effects in irreversible thermodynamics. The mechanism of turbulent cross-effects is completely different and is associated with turbulent transport in reacting flows. The analyzed effects may result in the excitation of a large-scale instability which causes formation of large-scale inhomogeneities in the spatial distributions of temperatures and number densities of admixtures. [S0031-9007(97)04944-2]

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The problem of turbulent transport in flows with chemical reactions (or phase transitions, e.g., evaporation or condensation) is of fundamental importance in view of various applications (e.g., physics of turbulent atmospheres of the Earth and planets, environmental physics, turbulent combustion). In recent years transport of passive scalar by turbulent fluid flow has been a subject of an active research. The nature of the intermittency and anomalous scalings [1,2], and the mechanisms of formation of inhomogeneous structures of scalar fields in turbulent fluid flows [3,4] were elucidated recently. In particular, the inhomogeneous structures in scalar fields (e.g., the number density of inertial particles or gases) are formed due to the effects of turbulent barodiffusion and turbulent thermal diffusion which cause additional turbulent nondiffusive fluxes of particles [3,4]. At large Reynolds and Peclet numbers these turbulent fluxes are much larger than the molecular fluxes. Inertial particles and gases are concentrated in the vicinity of the minimum of the mean temperature of the surrounding fluid.

However, turbulent transport in chemically reacting flows and flows with heterogeneous phase transitions remains poorly understood. In this Letter we show that turbulent diffusion can be strongly depleted by chemical reactions or phase transitions. We have found that there exist additional turbulent fluxes of number density of particles (effect of turbulent mutual diffusion of admixtures) and additional turbulent heat flux which is proportional to the gradient of number density of particles in flows with chemical reactions or phase transitions. Cross-effects (e.g., heat flux caused by concentration gradient, Dufour effect) are well known in irreversible thermodynamics (see, e.g., [5]). Many of them were discovered more than 100 years ago. In this Letter we show that similar effects can occur also in turbulent reactive flows. Certainly the mechanism of “turbulent”

cross-effects is completely different and is associated with turbulent transport in reacting flows.

Consider a mixture of reacting gases or droplets advected by a turbulent fluid flow. Reactions comprise either chemical reactions or heterogeneous phase transitions of the first kind (e.g., evaporation or condensation). It is assumed that densities of gaseous admixtures and spatial density of droplets ( $\rho_i \equiv n_i m_i$ ) are much smaller than the fluid density  $\rho_f$ , and that turbulent fluid flow is not affected by chemical reactions or phase transitions. Here  $m_i$  and  $n_i$  are the mass and number density of droplets, respectively. Temperatures of the fluid and gaseous admixtures (or droplets) are assumed to be the same. The number density  $n_i(t, \mathbf{r})$  of admixtures and the temperature field  $T_f(t, \mathbf{r})$  are determined by equations

$$\partial n_i / \partial t + \nabla \cdot (n_i \mathbf{v}_i) = \alpha_i I(n, T_f) + \nabla \cdot (\kappa_i \nabla n_i), \quad (1)$$

$$\begin{aligned} \partial T_f / \partial t + (\mathbf{v} \cdot \nabla) T_f + (\gamma - 1) T_f (\nabla \cdot \mathbf{v}) \\ = \beta I(n, T_f) + \nabla \cdot (\kappa_f \nabla T_f), \quad (2) \end{aligned}$$

where  $T_f$  is the fluid temperature, function  $\alpha_i I(n, T_f)$  is the source of gaseous admixtures (or droplets),  $n = (n_1, n_2, \dots, n_k)$  are the number densities of admixtures,  $\alpha_i$  are the stoichiometric coefficients with appropriate signs for chemical reactions (or  $\alpha = -1$  in the equation for evaporating droplets number density, and  $\alpha$  is a number of molecules in a droplet in the equation for vapor number density),  $\beta = \Delta E / \rho_f c_p$ ,  $\Delta E$  is the energy release (or absorption) in an elementary act (e.g., an amount of heat required for an evaporation of a droplet, or energy released or absorbed during a chemical reaction),  $c_p$  is the specific heat of fluid at constant pressure  $P_f = \rho_f \kappa_B T_f / m_\mu$ ,  $m_\mu$  is the mass of molecules of the fluid,  $\kappa_B$  is Boltzmann constant,  $\mathbf{v}_i$  is a random velocity field of admixtures which they acquire in turbulent fluid velocity field  $\mathbf{v}$ ,  $\kappa_i$  is the coefficient of molecular diffusion of

admixture,  $\kappa_f$  is the coefficient of molecular thermal conductivity,  $\beta I(n, T_f)$  is the rate of energy release, and  $\gamma$  is the specific heats ratio. We consider a low-Mach-number compressible turbulent fluid flow. The velocity  $\mathbf{v}$  of the fluid satisfies the continuity equation

$$\partial \rho_f / \partial t + \nabla \cdot (\rho_f \mathbf{v}) = 0. \quad (3)$$

The velocity of gaseous admixture equals the fluid velocity, whereas the velocity of particles (droplets)  $\mathbf{v}_i$  is determined from the equation of motion for a particle:  $d\mathbf{v}_i/dt = (\mathbf{v} - \mathbf{v}_i)/\tau_i + \mathbf{f}$ , where  $\tau_i$  is the characteristic time of momentum coupling between the particle and surrounding fluid (Stokes time),  $\mathbf{f} = \mathbf{F}/m_i$ , and  $\mathbf{F}$  is the external force. The fluid velocity  $\mathbf{v}$  is determined by Navier-Stokes equation. Equations (2) and (3) yield

$$\partial T_f / \partial t + (\mathbf{v} \cdot \nabla) T_f = \beta I(n, T_f) + \kappa_f \Delta T_f, \quad (4)$$

where we change notations  $\gamma^{-1} \beta \rightarrow \beta$  (see, e.g., [6]).

Consider homogeneous equilibrium with  $I = 0$ . This equilibrium can exist in a turbulent medium when  $\text{Pe} = u_0 l_0 / \kappa_f \gg 1$ , where  $l_0$  is the maximum scale of turbulent fluid motions,  $u_0$  is the characteristic turbulent fluid velocity in the scale  $l_0$ . In this case in the equilibrium  $\nabla \cdot \mathbf{v} \approx 0$ , because for low Mach numbers and  $I = 0$  the fluid velocity divergence  $\nabla \cdot \mathbf{v} \propto \text{Pe}^{-1} \Delta T_f / T_f$  (see, e.g., [6]). Now we study deviation from this equilibrium. Linearizing Eqs. (1) and (4) in the vicinity of the equilibrium we obtain a system of equations for small perturbations

$$\partial a^\alpha / \partial t + (\mathbf{v}_\beta^\alpha \cdot \nabla) a^\beta = P_\beta^\alpha a^\beta + \kappa_\beta^\alpha \Delta a^\beta + \Gamma^\alpha, \quad (5)$$

where the notation  $C_\beta^\alpha a^\beta$  assumes summation over  $\beta$ ,  $P_\beta^\alpha = h^\alpha (\partial I / \partial a^\beta)$ , and  $\mathbf{v}_\beta^\alpha = \mathbf{v}^\beta$ , if  $\alpha = \beta$  and  $\mathbf{v}_\beta^\alpha = 0$  if  $\alpha \neq \beta$ ; similarly  $\kappa_\beta^\alpha = \kappa^\beta$ , if  $\alpha = \beta$ ,  $\kappa_\beta^\alpha = 0$ , if  $\alpha \neq \beta$ ,

$$a^\alpha = \begin{pmatrix} n_{\{j\}} \\ T_f \end{pmatrix}, \quad \Gamma^\alpha = \begin{pmatrix} \Gamma_{\{j\}} \\ 0 \end{pmatrix}, \quad \mathbf{v}^\beta = \begin{pmatrix} \mathbf{v}_{\{j\}} \\ \mathbf{v} \end{pmatrix},$$

$$\kappa^\beta = \begin{pmatrix} \kappa_{\{j\}} \\ \kappa_f \end{pmatrix}, \quad h^\alpha = \begin{pmatrix} \alpha_{\{j\}} \\ \beta \end{pmatrix},$$

the notation  $c_{\{j\}}$  is the column of the elements  $c_1, c_2, \dots, c_k$ ,  $\Gamma_i = -n_i^{(0)} b_i$ ,  $b_i = \nabla \cdot \mathbf{v}_i^{(1)}$ ,  $\mathbf{v}_i^{(1)}$  are the perturbations of velocity, and  $\mathbf{v}_i$  is the velocity at the equilibrium.

We study the large-scale dynamics and average Eq. (5) over an ensemble of random velocity fluctuations. For this purpose we use the method of path integrals (see, e.g., [2-4,7,8]). The solution of Eq. (5) with an initial condition at  $t = t_0$  is given by a modified Feynman-Kac formula,

$$a^\alpha(t, \mathbf{x}) = M\{G_\beta^\alpha(t, t_0) a^\beta[t_0, \hat{\xi}(t, t_0)]\} + M\left\{\int_{t_0}^t G_\beta^\alpha(t, t') J^\beta[t', \hat{\xi}(t, t')] dt'\right\},$$

where  $\hat{\xi} \equiv \vec{\xi}_\beta^\alpha$  are Wiener paths,

$$\vec{\xi}_\beta^\alpha(t, t_0) = \mathbf{x} \delta_\beta^\alpha - \int_0^{t-t_0} \mathbf{v}_\beta^\alpha[t_s, \hat{\xi}(t, t_s)] ds + (2\kappa_\beta^\alpha)^{1/2} \mathbf{w}(t - t_0),$$

$\delta_\beta^\alpha = 1$ , if  $\alpha = \beta$ , and  $\delta_\beta^\alpha = 0$ , if  $\alpha \neq \beta$ ,  $t_s = t - s$ , and  $M\{\cdot\}$  denotes the mathematical expectation over the Wiener paths. Function  $G_\beta^\alpha$  is determined by the equation  $dG_\beta^\alpha/dt = P_\gamma^\alpha G_\beta^\gamma$  with the initial condition  $G_\beta^\alpha(t = t_0, t_0) = \delta_\beta^\alpha$ . The solution of this equation is given by  $G_\beta^\alpha = \delta_\beta^\alpha - \tau_c \{\exp[-(t - t_0)/\tau_c] - 1\} P_\beta^\alpha$ , where  $\tau_c = -\lambda^{-1}$  is the relaxation time related with chemical processes (or phase transitions),  $\lambda$  is the eigenvalue of the equations:  $P_\beta^\alpha W^\beta = \lambda W^\alpha$  and  $W_\alpha P_\beta^\alpha = \lambda W_\beta$ . Here the eigenvectors  $W^\alpha$  and  $W_\beta$  satisfy the identities  $P_\beta^\alpha = -W^\alpha W_\beta$  and  $W_\alpha W^\alpha = -\lambda$ . Therefore  $\lambda \equiv P_\alpha^\alpha = -\sum_{j=1}^k \tilde{\tau}_j^{-1}$  is the trace of the tensor  $P_\beta^\alpha$  and  $P_\gamma^\alpha P_\beta^\gamma = \lambda P_\beta^\alpha$ . Here  $\tilde{\tau}_j^{-1} = -\alpha_j (\partial I / \partial n_j) - \beta (\partial I / \partial T)$ .

The use of the technique of path integrals (see, e.g., [4]) allows one to derive equations for the mean fields  $A^\alpha$

$$\partial A^\alpha / \partial t + \vec{\nabla} \cdot (\mathbf{U}_\beta^\alpha A^\beta) = \vec{\nabla} \cdot (\hat{B}_\beta^\alpha \vec{\nabla} A^\beta) + P_\beta^\alpha A^\beta - \vec{\nabla} \cdot \mathbf{J}^\alpha, \quad (6)$$

where  $A^\alpha = \langle a^\alpha \rangle$ ,  $\mathbf{U}_\beta^\alpha = \mathbf{V}_\beta^\alpha + \sigma P_\gamma^\alpha \mathbf{V}_\beta^\gamma$ ,  $\hat{B}_\beta^\alpha = (\delta_\gamma^\alpha + \chi P_\gamma^\alpha) \hat{D}_\beta^\gamma + \kappa_\beta^\alpha \hat{\delta}$ ,  $\mathbf{J}^\alpha = \langle \tau \mathbf{u}_\beta^\alpha \Gamma^\beta \rangle$ ,  $\hat{\delta} = \delta_{mn}$ ,  $\hat{D}_\beta^\alpha = \langle \tau (\mathbf{u}_\gamma^\alpha)_m (\mathbf{u}_\beta^\gamma)_n \rangle$ ,  $\sigma = \tau_c \{1 + \tau_* [\exp(-2/\tau_*) - 1]/2\}$ ,  $\chi = \tau_c [1 - \tau_* - \tau_*^2 \ln(\tau_* + \text{Re}_*^s)]$ ,  $s = (q - 1)/(3 - q)$ ,  $\tau_* = \tau_c / \tau_0$ ,  $\tau_0 = l_0 / u_0$ ,  $\text{Re}_* = \min\{\text{Re}, \text{Pe}_i\}$ ,  $\text{Re} = l_0 u_0 / \nu$  is the Reynolds number,  $\nu$  is the kinematic viscosity,  $\text{Pe}_i = l_0 u_0 / \kappa_i$  are the Peclet numbers,  $\mathbf{v}_i = \mathbf{V}_i + \mathbf{u}_i$ ,  $\mathbf{V}_i = \langle \mathbf{v}_i \rangle$  are the mean particles velocities,  $\mathbf{u}_i$  are the fluctuations of the particles velocity,  $\mathbf{V}_\beta^\alpha = \langle \mathbf{v}_\beta^\alpha \rangle$ ,  $\mathbf{v}_\beta^\alpha = \mathbf{V}_\beta^\alpha + \mathbf{u}_\beta^\alpha$ , and  $\tau$  is the momentum relaxation time of random velocity field  $\mathbf{u}_i$ , which depends on scale of turbulent motion. We use here the dependence  $\tau(k) = \tau_0 (k/k_0)^{1-p}$ , where  $k$  is the wave number,  $k_0 = l_0^{-1}$ ,  $p$  is the exponent of the spectrum of the kinetic turbulent energy of fluid, and  $q = 2p - 1$ . We use here for simplicity a  $\delta$  correlated in time random process to describe a turbulent velocity field. However, the results remain valid also for the velocity field with a finite correlation time, if the mean number density of particles varies slowly in comparison with the correlation time of the turbulent flow. For simplicity here we consider the case  $\langle b_i \rangle = 0$ . Notably, Eq. (6) shows that an effective mean velocity ( $\mathbf{U}_\beta^\alpha$ ) of gaseous admixtures does not coincide with a mean fluid velocity ( $\mathbf{V}_\beta^\alpha$ ) due to chemical reactions or phase transitions.

The equation for the mean temperature reads

$$\partial T / \partial t = \nabla \cdot \left( B \nabla T + \sum_{k=1}^n \tilde{S}_k \nabla N_k \right) + \beta \sum_{k=1}^n (\partial I / \partial N_k)_0 N_k + \beta (\partial I / \partial T)_0 T, \quad (7)$$

$$B = \kappa_T[1 + \chi\beta(\partial I/\partial T)_0] + \kappa_f \quad (8)$$

[see Eq. (6)], where  $T = \langle T_f \rangle$  is the mean temperature, partial derivatives  $(\partial I/\partial N)_0$  and  $(\partial I/\partial T)_0$  are calculated at the equilibrium,  $\tilde{S}_k = \chi\beta(\partial I/\partial N_k)_0 D_k$ , and  $\kappa_T = l_0 u_0/3$  is the turbulent thermal conductivity. For simplicity, hereafter we consider the case of a zero mean velocity  $\mathbf{V} = 0$ . The term which is proportional to  $\nabla N_k$  in Eq. (7) can be interpreted as a turbulent analog of the effect of Dufour [5]; i.e., it describes turbulent transport of heat due to the number density gradient of chemically reacting admixtures. The second term in Eq. (8) describes an effect of chemical reactions (or phase transitions) on the turbulent heat flux. When  $\beta(\partial I/\partial T)_0 < 0$  the chemical reactions result in a reduction of the turbulent thermal conductivity.

Now we calculate the flux  $\mathbf{J}^\alpha$ . We use an equation  $\langle \tau \mathbf{u}_i b_i \rangle \approx \langle \tau \mathbf{u} b \rangle + (\tau_i v_T^2/T_*) \langle \tau \mathbf{u} \Delta \theta \rangle$  [3], where  $v_T^2 = \kappa_B T_0/m_\mu$ ,  $T_0$  is the temperature at the equilibrium,  $T_f = T + \theta$ ,  $\theta$  are fluctuations of temperature, and  $\mathbf{u}$  are fluctuations of velocity of the surrounding fluid. This equation follows from a solution of the equation of motion for particles with  $\rho_p \gg \rho_f$  and small Stokes time, i.e.,  $\mathbf{v}_i = \mathbf{v} - \tau_i(d\mathbf{v}/dt) + O(\tau_i^2)$  (see, e.g., [9]), where  $\rho_p$  is the material density of particles. Here we use the equation of state and Navier-Stokes equation for fluid, and an equation for turbulent thermal flux  $\langle \mathbf{u} \theta \rangle = -(B - \kappa_f)\nabla T - \sum_{k=1}^n \tilde{S}_k \nabla N_k$  which is determined from Eqs. (4) and (7). This yields the flux  $\mathbf{J}^\alpha$  and therefore Eq. (6) for a mean number density of the particles  $N_i$  is given by

$$\partial N_i/\partial t = \nabla \cdot \left( B_i \nabla N_i + \sum_{k=1, k \neq i}^n S_k \nabla N_k + W_i \nabla T \right) + \alpha_i \sum_{k=1}^n (\partial I/\partial N_k)_0 \tilde{z}_i N_k + \alpha_i (\partial I/\partial T)_0 T, \quad (9)$$

$$B_i = D_i [1 + (\partial I/\partial N_i)_0 \tilde{\zeta}_i] + \kappa_i, \quad (10)$$

where  $S_k = D_i (\partial I/\partial N_k)_0 \tilde{z}_i$ ,  $W_i = \kappa_T [(\xi_i + \eta_i)/T_0 + (\partial I/\partial T)_0 \tilde{\zeta}_i]$ ,  $\eta_i = 0$  for gaseous admixtures,  $\eta_i = (3N_i^{(0)}/\text{Pe})(m_i/m_\mu) \ln \text{Re}_*$  for particles [3],  $N_i^{(0)}$  is the mean number density at the equilibrium,  $\xi_i = N_i^{(0)} D_i/\kappa_T$ ,  $\tilde{\zeta}_i = \chi(\alpha_i D_k/D_i + \beta \eta_i/T_0)$ , and  $\zeta_i = \tilde{\zeta}_i (k = i)$ . The first term in Eq. (9) describes turbulent diffusion, and the third term determines turbulent thermal diffusion. These processes are modified by chemical reactions (or phase transitions). The second term in Eq. (9) corresponds to a mutual turbulent diffusion of admixtures caused by chemical reactions (or phase transitions) in turbulent fluid flow.

Note that when  $\alpha_i (\partial I/\partial N_i)_0 < 0$  the chemical equilibrium is stable. For gases  $\eta_i = 0$ . Thus it is seen from Eq. (10) that the "effective" diffusion coefficient  $B_i$  can be much less than the coefficient of turbulent diffusion  $D_i$  when  $|(\partial I/\partial N_i)_0 \tilde{\zeta}_i| \rightarrow 1$ . This implies that chemical reactions (or phase transitions) can result in a strong depletion of turbulent diffusion. Consider for simplic-

ity a reaction  $A \rightarrow C$  when the inverse reaction  $C \rightarrow A$  does not occur. The derivation of the equation for the mean fields by method of path integrals shows that when  $\tau \gg \tau_c$  the turbulent diffusion coefficient is given by  $B_i \sim \tau_c \langle \mathbf{u}^2 \rangle$  [see Eq. (10)]. Here  $\tau_c$  is the characteristic chemical time,  $\tau$  is the correlation time of the velocity field. This expression for the turbulent diffusion coefficient can be obtained by simple estimation. Indeed, we average linearized Eq. (1) over the ensemble of the turbulent velocity fluctuations and subtract the obtained averaged equation from Eq. (1). This yields an equation for the turbulent component  $\Theta$  of the number density of the reagent  $A$

$$\partial \Theta/\partial t - D \Delta \Theta = -(\mathbf{u} \cdot \nabla) N - R - \Theta/\tau_c,$$

where  $n_p = N + \Theta$ ,  $R = (\mathbf{u} \cdot \nabla) \Theta - \langle (\mathbf{u} \cdot \nabla) \Theta \rangle$ . For simplicity we consider here the case  $\nabla \cdot \mathbf{u} = 0$ . The magnitude of  $\partial \Theta/\partial t - D \Delta \Theta + R$  in inertial range can be estimated as  $\Theta/\tau$ . Thus the turbulent field  $\Theta$  is of the order of  $\Theta \sim -\tau_{\text{eff}} (\mathbf{u} \cdot \nabla) N$ , where  $\tau_{\text{eff}}^{-1} = \tau^{-1} + \tau_c^{-1}$ . Now we calculate the turbulent flux of reagent  $A$ :  $\mathbf{J}_T = \langle \mathbf{u} \Theta \rangle \sim -\tau_{\text{eff}} \langle \mathbf{u} u_j \rangle \nabla_j N$ . Since  $\tau \gg \tau_c$  we obtain that  $\tau_{\text{eff}} \sim \tau_c$ . Therefore the turbulent diffusion coefficient is given by  $B_i \sim \tau_c \langle \mathbf{u}^2 \rangle$ . Without chemical reaction the turbulent diffusion coefficient is given by  $B_0 \sim \tau \langle \mathbf{u}^2 \rangle$ . This means that  $B_i \ll B_0$ ; i.e., the turbulent diffusion coefficient is strongly reduced due to chemical reactions. The above estimation shows that depletion of turbulent diffusion cannot be described simply as an additive sink term in the equation for the mean number density.

The physics of this phenomenon is as follows. During transport of admixture  $A$  by a turbulent fluid flow the number density of the admixture  $A$  decreases due to the chemical reaction  $A \rightarrow C$  with a very short time  $\tau_c \ll \tau_0$ . Thus turbulent diffusion does not contribute to the mass flux of a reagent  $A$ . The turbulent mixing is effective only for the product of reaction  $C$ . The situation becomes more complicated for a multicomponent chemical reaction with the inverse reaction. The effect of depletion of the turbulent diffusion is similar to that for the reaction  $A \rightarrow C$ . The total chemical relaxation time  $\tau_c$  is determined by an equation  $\tau_c^{-1} = \sum_{j=1}^k \tilde{\tau}_j^{-1}$ , where  $\tilde{\tau}_j$  is the relaxation time of the  $j$  component. The components with the time  $\tau_i \gg \tau_c$  have the turbulent diffusion coefficient  $\sim B_0$ , whereas turbulent diffusion coefficients for the components with  $\tau_i \sim \tau_c$  are strongly reduced.

Now we discuss the mechanism of the mutual turbulent diffusion of admixtures. Note that the mutual molecular diffusion of admixtures is caused by interaction between gaseous admixtures due to collisions of molecules of the gaseous admixtures. In turbulent flow with chemical reactions inhomogeneities of the number density of one of the reagents causes fast change (on the chemical reaction time scale  $\tau_c$ ) of the number density of other components due to the shift from the chemical equilibrium. Similarly,

the inhomogeneities of the number density of the admixture cause heat release (or absorption) due to the thermal effects of the chemical reactions, i.e., additional nondiffusive turbulent heat fluxes. These fluxes can be interpreted as turbulent analogs of the known cross-effects in irreversible thermodynamics.

Now we show that the equilibrium state is unstable with respect to large-scale perturbations (in scales are much larger than the maximum scale of turbulent motions). The mechanism of the instability is as follows. Large-scale temperature perturbations of the mean temperature (e.g., mean temperature reduction in some region) result in accumulation of droplets in the regions with temperature minimum due to the effect of turbulent thermal diffusion. Evaporation of droplets causes further temperature decrease in this region. The latter results in the increase of the droplets number density and temperature reduction. Therefore the instability is excited. This simple mechanism is complicated by the effect of chemical reactions on turbulent transport: reduction of turbulent diffusion and turbulent analogs of cross-effects. Large-scale perturbations of the temperature  $T$  and the number density of admixtures  $N_i$  result in the appearance of the additional flows with  $\nabla \cdot \mathbf{v}^{(1)} \neq 0$ . It is conceivable to suggest that release (or absorption) of energy during exothermic (or endothermic) chemical reactions in the vicinity of the large-scale inhomogeneities increases, and the temperature of surrounding fluid increases (or decreases). Therefore the large-scale perturbations of the temperature and the number density of admixtures can grow, and under certain conditions the large-scale instability can be excited. This instability causes formation of large-scale inhomogeneities of the temperature and number density of droplets and gaseous admixtures.

Now we study this instability which is described by Eqs. (7) and (9). Consider an example of phase transition, e.g., the evaporation of droplets. In the case of the evaporation of droplets  $\eta \gg \xi$ . The source function  $I$  for the phase transition is given by  $I = f(T)N_0 - I_*$ , and  $\alpha = -1$ , where  $I_*$  is the external source of the droplets, and function  $f(T)$  is determined by the evaporation time of the droplets. The growth rate of the large-scale perturbations of the temperature and droplets number density is given by  $\gamma_e = (\beta_* \eta_* \Lambda - 1)k^2 D_T$ , where  $\eta_* = \eta/N_0$ ,  $N_0$  is the droplets number density at the equilibrium,  $\Lambda = (\partial I / \partial N)_0 \tau_c$ ,  $\beta_* = -\beta N_0 / T_0 = \mu Q \rho_d / (\gamma^2 E_T \rho_f)$ , the droplets spatial density  $\rho_d = m_d N_0$ ,  $m_d$  is the mass of a droplet,  $Q$  is the latent heat of evaporation per one mole of water,  $E_T$  is the thermal energy per mole, and  $\mu$  is

the molecular weight of the surrounding fluid. For example, when  $\rho_d \sim 10^{-1} \text{ g/m}^3$ ,  $T \sim 300 \text{ K}$ , and  $\text{Re} \sim 10^5$  we find that the minimum size of droplets for which the instability is excited must be larger than  $20 \text{ }\mu\text{m}$ . This instability is completely different from that studied in [3,4] since it is accompanied by the change of the temperature distribution. In addition, this instability strongly depends on the latent heat  $Q$  of the phase transition, and it can be excited even for homogeneous distribution of the equilibrium mean temperature of fluid. On the other hand, the instability studied in [3,4] is not excited when fluid temperature is homogeneous.

The similar instability can exist in chemically reacting flows. However, the mechanism of the instability is complicated by the additional factors, e.g., photochemical and electrochemical processes. These effects are the subject of an ongoing study.

In summary, coupling between chemical reactions (or phase transitions) and turbulent transport yields a number of new effects: depletion of turbulent diffusion and turbulent analogs of cross-effects known in irreversible thermodynamics. The discovered effects have a strong influence on the dynamics of turbulent mixing.

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