KINETICS OF PHASE TRANSFORMATIONS ON ISOTHERMAL SLOT CHANNEL WALLS

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The determination of the numerical density of molecules flying off from slot channel walls of different geometric shape whose internal surfaces are covered by a sublimable material is reduced to the solution of the Helmholtz equation. Simple formulas are obtained for computation.

The asymptotic of rarefied gas flow of Hill–Shaw type in the gap formed by parallel walls located at a distance H whose scale L in the plane of symmetry of the gap is quite large as compared with the mean free path length of the molecules ($Kn_L = \lambda/L << 1$) is constructed in [1]. The flow of vapor in the gap occurring as a result of sublimation (evaporation) or condensation on the internal surface of slot channel walls to which the necessary quantity of heat is supplied from outside (eliminated) is considered as an illustration. The problem was solved in a linear approximation, which corresponds to "slow" phase transition processes [2], when the macroscopic velocity of the vapor along the normal to the interphasal surface compared with the mean thermal velocity of the molecules and the relative temperature change are small.

Analysis of phase transformation on slot channel isothermal walls (T_w = const, $\tau_w = 0$) from the formal viewpoint is simpler than elucidated in [1]. In this case $n_{ew} = n_{ew}(T_w)$ = const and, consequently, a homogeneous Helmholtz equation can be written at once from (19) and (20) obtained in [1] (without additional linearization) for the new independent variable $\delta n_w = n_w - n_{ew}$ which characterizes the degree of nonequilibrium of the process being studied

$$\Delta\delta n_w - c_4^2\delta n_w = 0,$$
$$c_4 = \gamma_0/\varepsilon;\quad \gamma_0^2 = \frac{-\beta}{(1-\beta)\sqrt{\pi}\,Q_v^{(0)}(\alpha)}. \tag{1}$$

In contrast to (22) from [1], this equation always has a negative coefficient ($-c_4^2$) before the δn_w, which somewhat simplifies the investigation of appropriate problems (a single mode of solution in the whole range of variation of the physical parameters; it is not necessary to pose additional conditions of the radiation condition type in the analysis of phase transitions in an infinite slot channel). At the same time, attention should be turned to the fact that this equation is singularly perturbed if only the condensation coefficient β characterizing the fraction of molecules captured by the phase transition surface out of the whole number contained in the incident stream is not a quantity of the same order of smallness as the parameter $\varepsilon = H/L << 1$. Therefore, for $\beta >> \varepsilon$ ($c_4 >> 1$) a perceptible change in the numerical density of the molecules flying out from the channel walls will occur only in a comparatively narrow band, adjoining the slot channel exit and being an original "boundary" layer in whose computation the one-dimensional analog of (1) can be utilized ($\Delta = d^2/d\xi^2$; ξ is measured along the normal to the open part of the slot channel contours or the wall domains in which phase transformations occur). The width of this band is $b_* = O(H/\gamma_0) = O(H/\beta)$; consequently, (1) has meaning only for $\beta < 1$ since otherwise the asymptotic of the Hill–Shaw type flow constructed under the assumption that the linear flow scale in the plane of the channel is many times greater than its height $H/L << 1$ [3] is not applicable in the band mentioned. This constraint is substantial: the theory elucidated here is not applicable to the solution of problems on phase transformations on isothermal slot channel walls with insufficiently small condensation coefficients.

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If the condition $\beta \ll 1$ is satisfied, then (1) is applicable, and all methods discussed in [1] are suitable for its solution, taking into account the above-mentioned simplifying analyses of the circumstances. All the solutions presented in [1] are converted, without difficulty, into solutions of the problems considered here that have the same geometry. Thus, for instance, for an open gap between parallel rectangular plates ($-1 \leqq \xi \leqq 1$, $-\eta_0 \leqq \eta \leqq \eta_0$, $b/L = \eta_0$) the formula characterizing the distribution n_w of the numerical density of the molecules, flying off from the walls, over the gap has the form

$$\delta n_w = \delta n_w(\Gamma)\left[F_0(\xi, c_4) + \sum_{n=0}^{\infty} \frac{2(-1)^n c_4^2}{\mu_n(\mu_n^2 + c_4^2)} \frac{\mathrm{ch}(\sqrt{\mu_n^2 + c_4^2}\,\eta)}{\mathrm{ch}(\sqrt{\mu_n^2 + c_4^2}\,\eta_0)} \cos(\mu_n \xi)\right]. \tag{2}$$

Here $\delta n_w(\Gamma) = n_w(\Gamma) - n_{ew}(T_w) = \mathrm{const}$, $n_w(\Gamma)$ is determined by the pressure at the slot channel exit, $\xi = \pm 1$, $\eta = \pm\eta_0$, $[P_\Gamma(\Gamma) = mn_w(\Gamma)RT_w]$; $\mu_n = \pi(n + 1/2)$; $F_0(\xi, c_4) = \mathrm{ch}(c_4\xi)/\mathrm{ch}(c_4)$.

As regards the other solutions presented in [1], they are complicated somewhat in this paper: we shall consider that the phase transformations do not hold on the whole internal surface of the channel walls. Such a formulation of the problem is of interest, say, for the analysis of the sublimation of a thin coating deposited preliminarily on the whole wall surface or on part. For the two examples examined in [1], the solutions (open on two sides ($\xi = \pm 1$) planar slot channel ($\ell = 0$), and the gap between two discs of radius $\xi = 1$ ($\ell = 1$)) we assume that the phase transformations occur only in the domain $|\xi| \leq \xi' < 1$. Then by analogy with (24) from [1] we can write for the points $\xi_* \leq \xi_*'$:

$$\delta n_w(\xi_*) = \frac{F_l(\xi_*, 1)}{F_l(\xi_*', 1)} \delta n_w(\xi_*'), \quad \xi_* = c_4\xi, \quad \xi_*' = c_4\xi'. \tag{3}$$

The second forms of (24), presented in [1], correspond here naturally to the functions $F_\ell(\xi_*, 1)$ since $(-c_4^2) < 0$:

$$F_0(\xi_*, 1) = \frac{\mathrm{ch}(1, \xi_*)}{\mathrm{ch}(1)}, \quad F_1(\xi_*, 1) = \frac{I_0(1, \xi_*)}{I_0(1)},$$

where $I_0(x)$ is the Bessel function of the imaginary argument.

For the domains $\xi' < |\xi| < 1$ within which phase transformations do not occur, a relationship can be written that characterizes the continuity condition for the vapor stream during stationary progress of the process

$$\begin{gathered} \xi^l \frac{d\delta n_w}{d\xi} = K_l = \mathrm{const}, \quad (\delta n_w(\Gamma) = \delta n_w(1)), \\ K_0 = \frac{\delta n_w(\Gamma) - \delta n_w(\xi')}{1 - \xi'}, \quad K_1 = \frac{\delta n_w(\Gamma) - \delta n_w(\xi')}{-\ln \xi'}. \end{gathered} \tag{4}$$

But

$$\begin{gathered} K_0 = \frac{d\delta n_w}{d\xi|_{\xi \to \xi' - 0}} = c_4 \,\mathrm{th}(c_4\xi')\, \delta n_w(\xi'), \\ K_1 = \xi' \frac{d\delta n_w}{d\xi|_{\xi \to \xi' - 0}} = c_4\xi' \frac{I_1(c_4\xi')}{I_0(c_4\xi')} \delta n_w(\xi'). \end{gathered} \tag{5}$$

Therefore, in the domain $|\xi_*| < \xi_*'$ ($\xi_* = c_4\xi$)

$$\begin{gathered} \delta n_w(\xi_*) = \delta n_w(\Gamma)\, \Phi_l(\xi_*, \xi_*', c_4), \\ \Phi_0(\xi_*, \xi_*', c_4) = \frac{\mathrm{ch}\,\xi_*}{\mathrm{ch}\,\xi_*' + (c_4 - \xi_*')\,\mathrm{sh}\,\xi_*'}, \\ \Phi_1(\xi_*, \xi_*', c_4) = \frac{I_0(\xi_*)}{I_0(\xi_*') + \xi_*' \ln(c_4/\xi_*')\, I_1(\xi_*')}. \end{gathered} \tag{6}$$

An expression for the numerical density of molecules flying off from the walls in the domains $\xi^0 < \xi'' \leqq \xi \leqq \xi' < 1$ of the gap between parallel annular plates ($\xi^0 \leqq \xi \leqq 1$) can also be obtained in an analogous manner when the circles $\xi = \xi''$ and $\xi = \xi'$ correspond to the boundaries of the phase transition surface. In this case (3) takes the form

$$\delta n_w(\xi_*) = I_0(\xi_*, \xi_*'')\delta n_w(\xi_*') - I_0(\xi_*, \xi_*')\,\delta n_w(\xi_*''),$$

$$I_0(\xi_*, \xi_*'^{('')}) = \frac{I(\xi_*, \xi_*'^{('')})}{I(\xi_*'', \xi_*')}, \quad I(x, y) = I_0(x)K_0(y) - I_0(y)K_0(x),$$

$$\delta n_w(\xi_*'') = \delta n_w(\Gamma)\frac{z_1}{z_2}, \tag{7}$$

$$z_1 = \frac{1}{\xi_*'' \ln(\xi_*''/\xi_*^0)} + \frac{I_0'(\xi_*'', \xi_*'')}{\xi_*' \ln(\xi_*'/c_4)\, I_0'(\xi_*', \xi_*'') - 1},$$

$$z_2 = \frac{1}{\xi_*'' \ln(\xi_*''/\xi_*^0)} - \frac{\xi_*' \ln(\xi_*'/c_4)\,[I_0'(\xi_*', \xi_*'')\,I_0'(\xi_*'', \xi_*') - I_0'(\xi_*', \xi_*')\,I_0'(\xi_*'', \xi_*'')] - I_0'(\xi_*'', \xi_*')}{\xi_*' \ln(\xi_*'/c_4)\, I_0'(\xi_*', \xi_*'') - 1}, \tag{8}$$

$$\delta n_w(\xi_*') = \frac{\xi_*' \ln(\xi_*'/c_4)\, I_0'(\xi_*', \xi_*')\,\delta n_w(\xi_*'') - \delta n_w(\Gamma)}{\xi_*' \ln(\xi_*'/c_4)\, I_0'(\xi_*', \xi_*'') - 1},$$

$$I_0'(x, y) = \frac{I'(x, y)}{I(\xi_*', \xi_*'')}; \; I'(x, y) = I_1(x)K_0(y) - I_0(y)K_1(x).$$

The expressions (6) and (7) can be used, say, to determine the time of vacuum sublimation of a thin coating deposited on the internal surfaces of slot channel walls if the process proceeds quasistationarily, i.e., if the following condition is satisfied

$$\frac{1}{n_w}\,\frac{\partial \delta n_w}{\partial t} \ll \frac{|\nabla \langle \bar{u} \rangle|}{L}\sqrt{2RT_w}. \tag{9}$$

The angular brackets denote taking the average in velocity space. In the two examples examined above, the solutions follow from the expression (6)

$$\frac{\partial \delta n_w}{\partial t} = \delta n_w(\Gamma)\,\frac{\partial \Phi_l(\xi_*, \xi_*', c_4)}{\partial \xi_*'}\,\frac{d\xi_*'}{dt} \tag{10}$$

and the quasistationarity condition is written in the form

$$\frac{L}{\sqrt{2RT_w}}\,\frac{d\xi_*'}{dt} \ll \frac{\partial \langle u \rangle (\xi_*, \xi_*')/\partial \xi_* + (l/\xi_*)\langle u \rangle (\xi_*, \xi_*')}{\partial \Phi_l(\xi_*, \xi_*', c_4)/\partial \xi_*}.$$

By virtue of (13) from [1], it will be satisfied if

$$\frac{\beta}{1-\beta}\,\frac{m\delta n_w H}{\rho_0 \delta_0}\,\frac{\delta n_w}{n_w} \ll \left(\frac{H}{L}\right)^2 = \varepsilon^2.$$

Here ρ_0, δ_0 are the density and initial thickness of the subliming coating.

In the problems under consideration $\beta = O(\varepsilon)$. Therefore, for quasistationary progress of the process it is necessary that the surface density of the coating $\rho_0\delta_0$ be many times greater than $m\delta n_w H/\varepsilon$. But $m\delta n_w H$ is a quantity commensurate with the surface density of the sublimate in the slot channel, and consequently, the formulated condition is satisfied at the low pressures corresponding to the regimes being studied.

In the general case, the study of the kinetics of quasistationary sublimation of a thin layer on the internal walls of a slot channel reduces to the solution of sufficiently complex nonlinear and nonstationary functional equations. An interesting feature of the examples in which δn_w is determined by (6) is the fact that the quasistationary process of vacuum sublimation of a coating which has been deposited by a layer of thickness $\delta_0 \ll H$ on the channel wall internal surfaces will proceed self-similarly for them. Indeed, by using (19) from [1] and (6) for the vapor flux density distribution on the wall surfaces on which there is still a coating at the time t, an expression can be written

$$j_l(\xi_*) = j_m \Phi_l(\xi_*, \xi_*', c_4), \quad j_m = \frac{m\beta}{1-\beta}\sqrt{\frac{RT_w}{2\pi}}\,\delta n_w(\Gamma). \tag{11}$$

Therefore, we obtain for two arbitrary points characterized by the dimensionless coordinates ξ_{*1} and ξ_{*2}

$$\frac{j_l(\xi_{*1})}{j_l(\xi_{*2})} = \frac{\omega_l(\xi_{*1})}{\omega_l(\xi_{*2})}, \qquad \begin{aligned} \omega_0(\xi_*) &= \operatorname{ch}(\xi_*); \\ \omega_1(\xi_*) &= I_0(\xi_*). \end{aligned} \tag{12}$$

Naturally points are considered here that lie on the phase transformation surface, i.e., $|\xi_{*n}| \leqq |\xi_*'|$. The coordinate $\xi_*'(t)$ of the boundary of part of the internal wall surface having the coating varies in time as the boundary advances deep into the slot channel. The expression (12) means that sublimation from the phase transformation surface always occurs according to an identical law independently of the location of the boundary $\xi_*'(t)$ at this time t, more exactly, the curve $j_\ell(\xi_*, \xi_*')$ is transformed affinely into the curve

$$j_l[\xi_*, \xi_*'(t_2)] = j_l[\xi_*, \xi_*'(t_1)]\,\psi[\xi_*'(t_1), \xi_*'(t_2)] \tag{13}$$

as the parameter ξ_*' changes.

After the phase transformation boundary has been displaced deep into the slot channel, zero coating thickness will always correspond to it, i.e., the following condition will be satisfied

$$\lim_{\xi_* \to \xi_*'(t)} \delta[\xi_*, \xi_*'(t)] = 0, \tag{14}$$

and this means that a coating layer of thickness δ_0 will be carried away at points of the surface having the coordinate $\xi_*'(t)$ up to the time t, while at points with the coordinate $|\xi_*| < \xi_*(t)$ a layer of thickness $\delta_0 - \delta[\xi_*, \xi_*'(t)]$ will be carried away, consequently, taking (12) into account

$$\frac{\delta_0 - \delta[\xi_*, \xi_*'(t)]}{\delta_0} = \frac{\omega_l(\xi_*)}{\omega_l[\xi_*'(t)]}. \tag{15}$$

Therefore, in the two examples under consideration, after the beginning of boundary movement deep into the slot channel, the coating thickness profile will be described by the universal dependence

$$\delta(\xi_*, \xi_*') = \delta_0\left[1 - \frac{\omega_l(\xi_*)}{\omega_l(\xi_*')}\right]. \tag{16}$$

This latter circumstance simplifies the determination of the law of phase transformation front motion along the channel walls so much so that it permits reduction of the problem under consideration to the following elementary problem. The condition

$$\frac{d\delta}{dt} = \frac{\partial\delta}{\partial t} + \frac{\partial\delta}{\partial\xi_*|_{\xi_*=\xi_*'}}\,\frac{d\xi_*'}{dt} = 0 \tag{17}$$

should be satisfied on the moving boundary $\xi_*'(t)$ by virtue of (14), and therefore, the rate of phase transformation front displacements is

$$\frac{d\xi_*'}{dt} = -\left(\frac{\partial\delta/\partial t}{\partial\delta/\partial\xi_*}\right)_{\xi_*=\xi_*'}. \tag{18}$$

Using (11) and (16), we obtain

$$\frac{\partial\delta}{\partial t} = -\frac{j[\xi_*, \xi_*'(t)]}{\rho_0} = -\frac{j_m}{\rho_0}\Phi_l[\xi_*, \xi_*'(t), c_4], \tag{19}$$

$$\frac{\partial\delta}{\partial\xi_*} = -\delta_0\frac{\omega_l'(\xi_*)}{\omega_l(\xi_*')}, \qquad \begin{aligned} \omega_0'(\xi_*) &= \operatorname{sh}(\xi_*), \\ \omega_1'(\xi_*) &= I_1(\xi_*). \end{aligned} \tag{20}$$

Therefore taking (18) into account, we can write for the dimensionless time $\vartheta = (j_m t)/(\rho_0\delta_0)$

$$\frac{d\vartheta}{d\xi_*'} = -\frac{\omega_l'(\xi_*')}{\omega_l(\xi_*')}\psi_l(\xi_*', c_4), \quad \psi_l(\xi_*', c_4) = \frac{1}{\Phi_l(\xi_*', \xi_*', c_4)}.$$

If the dimensionless coordinate ξ_*^0 corresponds to the initial position of the coating boundary, then the time of boundary displacement until cancellation of the whole coating layer in the two cases under consideration will be determined by the expression

$$\vartheta_l = \int_0^{\xi_*^0} \psi_l(\xi_*, c_4)\frac{\omega_l'(\xi_*)}{\omega_l(\xi_*)}d\xi_*. \tag{21}$$

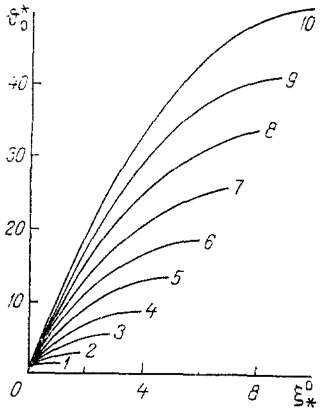

Fig. 1

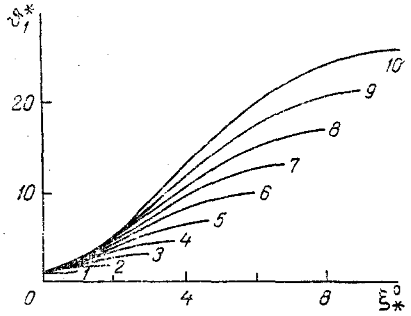

Fig. 2

Fig. 1. Total time of sublimation of a plane slot channel wall coating as a function of the initial position of the coating boundary: curves 1, 2, ..., 10 correspond to the values c_4 = 1, 2, ..., 10.

Fig. 2. Total time of sublimation of an axisymmetric slot channel coating as a function of the initial position of the coating boundary: curves 1, 2, ..., 10 correspond to the values c_4 = 1, 2, ..., 10.

The last integrals can be expressed in terms of elementary and cylindrical functions

$$\vartheta_0(\xi_*^0,\ c_4) = \frac{1}{2}(\xi_*^0)^2 + (c_4 - \xi_*^0)(\xi_*^0 - \mathrm{th}\,\xi_*^0),$$
$$\vartheta_1(\xi_*^0,\ c_4) = \frac{1}{4}(\xi_*^0)^2 + \xi_*^0 \ln\left(\frac{c_4}{\xi_*^0}\right)\left[\frac{\xi_*^0}{2} - \frac{I_1(\xi_*^0)}{I_0(\xi_*^0)}\right]. \tag{22}$$

The time preceding the beginning of front motion, i.e., the time of coating sublimation along the initial line of the front ($\xi_* = \xi_*^0$), must be appended to the time intervals formed above that correspond to phase transformation front advancement to the middle of the plane channel open on two sides (ϑ_0) or to the center of an axisymmetric (ϑ_1) channel. By virtue of (19) the dimensionless times $\vartheta_{\ell 1}$ determined by the expressions

$$\vartheta_{01} = 1 + (c_4 - \xi_*^0)\,\mathrm{th}\,\xi_*^0; \quad \vartheta_{11} = 1 + \xi_*^0 \ln\left(\frac{c_4}{\xi_*}\right)\frac{I_1(\xi_*^0)}{I_0(\xi_*^0)} \tag{23}$$

correspond to this period in the two cases under consideration. The total time of coating sublimation is determined by the sums $\vartheta_\ell^* = \vartheta_\ell + \vartheta_{\ell 1}$:

$$\vartheta_0^* = 1 + c_4\xi_*^0 - \frac{1}{2}(\xi_*^0)^2,$$
$$\vartheta_1^* = 1 + \frac{1}{2}\left[\ln\left(\frac{c_4}{\xi_*^0}\right) + \frac{1}{2}\right](\xi_*^0)^2. \tag{24}$$

The results of calculations using the last formulas are presented in Figs. 1 and 2 for a plane channel and the gap between discs.

If the internal surface is initially coated entirely by a subliming layer, i.e., $\xi_*^0 = c_4$, then the sublimation time will depend quadratically on the extent of the plane channel or the disc radius since in this case

$$\vartheta_0 = 1 + \frac{1}{2}c_4^2, \qquad \vartheta_1 = 1 + \frac{1}{4}c_4^2. \tag{25}$$

For very low values of the coefficient β and not too small values of the parameter ε, when $c_4 \ll 1$ according to (1), the total coating sublimation time is determined in practice by just the first stage ($\vartheta_{\ell 1}$) since the sublimation tempo in the whole extent of the slot channel is almost identical and limited only by the phase resistance of the subliming surface: the hydraulic resistance to the sublimate flowing in the gap is negligibly small. As is seen from (24), in this case

$$\vartheta_l^* = 1 + O(c_4^2).$$

The coating sublimation process for annular plates forming a doubly connected slot channel (in planform) is not self-similar and it is impossible to write a simple formula of the type (24) or (21) to compute the sublimation time. However, even in this case the kinetics can be computed comparatively simply by numerical methods since the internal and external phase transformation fronts also remain circles here that move oppositely to each other until they meet, which means completion of the process. For instance, a sufficiently simple algorithm can be proposed that will reduce to a step-by-step process of the following kind: we separate the domain $\xi'' < \xi < \xi'$ having the coating into N annular layers $\xi_n < \xi < \xi_{n+1}$, $\xi_n = \xi'' + (n/N)(\xi' - \xi'')$ and we calculate the change in coating thickness at the middle of each annular layer by formulas similar to (19), i.e., for $\xi = \xi_{n1} = {}^1/_2(\xi_n + \xi_{n+1})$ the functions $\delta(\xi_{n1}, t)$ are calculated. After the coating thickness has become zero, at the middle of one of the external annular layers, the appropriate front is displaced one step into the domain under consideration, i.e., continuous phase transformation front motion is here replaced by a stepwise process. It is natural to expect that the error in determining the coating sublimation time by such a numerical method will tend to zero as $N \to \infty$.

NOTATION

H and L, height and linear scale of the flow in the middle plane of a slot channel, respectively; λ, mean free path length of the molecules; $\varepsilon = H/L$; T, local temperature; $\tau = (T - T_0)/T_0$; n, numerical molecule density; $n_{\ell w}(T_w)$, equilibrium value of the numerical molecule density at the wall temperature T_w (on the saturation line); $\nu = (n - n_0)/n_0$; β, condensation coefficient; R, universal gas constant; m, molecule mass; $\bar{u}$, velocity; t, time; $Q_\nu^{(0)}(\alpha)$, dimensionless velocity (flow rate) averaged over the channel height; $\alpha = \sqrt{\pi}H/3\lambda$; j_m, vapor mass density from the channel wall; ξ, η, dimensionless rectangular coordinates in the middle plane of the channel, referred to the channel height; and Kn, Knudsen number. Subscripts: w, on the wall; and 0, at the point $\bar{r}_0$.

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OXYGEN-FREE VAPOR CONVERSION OF METHANE

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A reactor for the purpose of methane conversion is described. Comparison of a calculation of a mathematical model of the device with experimental data shows good agreement.

At the present time methane conversion is the basic industrial method for production of technological gases used to extract iron from ores by the direct reduction method. Tube furnaces used for this purpose require a large amount of expensive special fire-resistant steel, while mine reactors generate expensive gas due to their use of oxygen.

The reactor with a retarded fluidized bed described in [1] is free of these shortcomings. A diagram of this device is shown in Fig. 1. The retort 14 is packed with a catalyst 5, in the pores of which a fine grain material which acts as an intermediate heat-exchange agent circulates. In the "boiling" layer of these particles above the packing 7 a tuyere consisting of tube 9, "sleeve" 10, and perforations 8 is immersed.

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