Pergamon

Int. J. Heat Mass Transfer. Vol. 41, No. 16, pp. 2529–2536, 1998

Printed in Great Britain
0017-9310/98 \$19.00+0.00

PII: S0017-9310(97)00188-9

Several aspects of the return flows formation in horizontal CVD reactors

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(*Received* 8 *July* 1997)

Abstract—An explanation of the return flow onset behavior in a long horizontal CVD reactor is proposed on the basis of combined analytical and numerical approach. This approach allows one to obtain the condition of vortex onset in the form, that includes not only Grashof and Reynolds numbers, but also the temperature difference at the reactor top and bottom walls. Also some three-dimensional effects are investigated, which accompany the return flow formation in a horizontal CVD reactor. © 1998 Elsevier Science Ltd. All rights reserved

INTRODUCTION

The scientific interest to the gas recirculation in the reaction zone of CVD reactors is caused by at least two reasons. First, because of the delay in switching from one active component source to another when depositing III–V or II–VI compounds. Some amount of a reagent always accumulates in the region of vortex and remains there for a certain period of time after the supply of the reagent is terminated. To obtain abrupt layers one should wait until all molecules of the previous reagent leave the reactor chamber, before starting the supply of a new one. Another reason is that the transport processes in a gas change drastically when potential flow turns into a vortical one. The symmetry may be broken, so that the task dimensionality rises and much more complicated models are to be used.

MECHANISM OF THE RETURN FLOW FORMATION IN HORIZONTAL 2D REACTOR

Recirculation in CVD reactors has been the subject of a number of investigations, both theoretical and experimental. Among recent works devoted to this problem there is one published by Einset *et al.* [1]. It contains an attempt of theoretical explanation of the return flows formation. The authors proposed an approach to the problem and derived speculatively the condition of the return flow onset, which had been earlier obtained experimentally [2]. They tried to deduce the condition of the return flow formation from the tilt angle of the gas pressure isolines. The isobars' tilt to the axial reactor axis itself can not cause a recirculation, however the correlation between these two phenomena appeared to exist. To find the reason for this fact we will try to make some deeper insight into the mechanism of the return flow formation. Also, the question of how the condition of the recirculation onset depends on the temperature change in the reactor is stated for the first time, and the explanation for the empirically found condition of the vortex formation is proposed in this letter.

Let us consider the steady compressible flow at some low Reynolds number ($Re \ll 1$) in a horizontal reactor, which length L is much greater than the height h. Let the bottom wall, for certainty, to be hot: starting from the certain value of the horizontal coordinate x_0 its temperature is T_S. The top wall will be assumed cold (its temperature T_0 is maintained constant). Gas which comes into the reactor has the same temperature T_0. Let also both inlet and outlet to be located far enough from the point x_0, so that the flow far from x_0 up- and downstream of this point is 'fully developed' (does not depend on the horizontal coordinate). Also, some transition region presents inside the reactor, where the temperature and velocity fields change along X (horizontal) axis. This region occupies the part of the reactor space around x_0, and its length depends on the flow characteristics (Reynolds, Prandtl numbers and the relative temperature difference in the chamber).

It can be easily shown, that outside the transition region the velocity profile is a Poiseuille one (if ever the temperature dependence of the viscosity coefficient is not taken into account). The axial component of P'‡ gradient has a constant value there. The only

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‡ P' is not the gas pressure in its common sense, but the difference between the pressure in the reactor and the gas hydrostatic pressure for some arbitrary value of the temperature, and so, mixing of the concepts 'pressure' and 'P' may cause uncertainties. For instance, the direction of the P' vertical gradient component may be changed to opposite by choosing the appropriate value of the normalizing temperature. To avoid such uncertainties, we use notation 'P'' instead of 'pressure'.

NOMENCLATURE

C_p	gas heat capacity
Fr	Froude number
Gr	Grashof number
g	weight acceleration
$\mathbf{I}$	unit tensor
L_t	transition region length
P'	difference between the gas pressure and a hydrostatic pressure
Pr	Prandtl number
Re	Reynolds number
T	temperature
T_0	temperature of the cooled wall
T_S	temperature of the hot wall
T'	dimensionless temperature
U	velocity horizontal component
$\mathbf{V}$	gas velocity
V	velocity vertical component
V_0	velocity scaling factor
X	horizontal coordinate
x_0	starting point of the wall heated part
Z	vertical coordinate.

Greek symbols

β	adjusting parameter
λ	thermal conductivity
μ	viscosity
θ	relative temperature change $(T_S - T_0)/T_0$
ρ	gas density
ρ_0	density scaling factor.

difference between the flows far up- and downstream of x_0 is that the vertical component of the temperature gradient is zero and P' gradient is parallel to X axis in the region upstream of x_0, while in the flow downstream of x_0 the temperature vertical gradient has some finite value and P' gradient is tilted with respect to both X and Z (vertical) axes. Dimensionless form of the horizontal P' gradient for the Poiseuille flow is:

$$\frac{\partial P'}{\partial x} = -\frac{12}{Re} \tag{1}$$

where P' is scaled on the value $\rho_0 V_0^2$ (ρ_0 is the density scaling factor, V_0 is the velocity one), Re is the Reynolds number, the coordinates are normalized on the reactor height. (All other formulae are also presented in a dimensionless form.) The vertical P' gradient downstream of the transition region is given by:

$$\frac{\partial P'}{\partial z} = \frac{Gr}{Re^2\theta}\frac{T'}{1+T'} \tag{2}$$

where θ is the relative temperature change in the reactor $(T_S - T_0)/T_0$, T' is the dimensionless temperature $(T-T_0)/T_0$, Gr denotes the Grashof number.

Far upstream of the transition region $P'(x,z)$ dependence has asymptotically plane form. The plane is inclined to X axis at the angle given by equation (1). The shape of $P'(x,z)$ far downstream of the transition region may be found using equations (1) and (2). This geometry is sketched in Fig. 1. The transition region is filled by a pattern. For the sake of easily interpreting the downstream asymptotics of $P'(x,z)$ is shown plane. The disposition of the plane ABC and the surface EFGH (see Fig. 1) defines the onset of the recirculation in the following way: the return flow arises if the value of P', P_H (subscript letter means the corresponding point in Fig. 1) becomes greater than P_C, in other words, if positive horizontal component of the P' gradient exists (it is quite noticeable in Fig. 3, or Fig. 4 of [1]). If there is no heating ($\theta = 0$) P' gradient has only the horizontal component, and during any length l, P' drops by the value:

$$P_E - P_B = -\frac{12}{Re}l \tag{3}$$

The influence of the temperature gradient on the pressure drop in the transition region of length L_t may be accounted for by introducing an additional term in equation (3):

$$P_E - P_B = -\left(\frac{12}{Re}L_t + \Delta P_t\right) \tag{4}$$

where ΔP_t depends on the Froude number ($Fr = Re^2\theta/Gr$), since it is the Froude number, which represents in the only term of Navier–Stokes equations containing temperature [see equations (8) and (9) below] and also on θ.

To verify our speculations and to obtain an information about $\Delta P_t(Fr,\theta)$ dependence, series of calculations (equations and details of the numerical scheme are given in the Appendix) were performed for the reactor with high aspect ratio ($L/h = 15$).

The calculations, that covered the wide range of temperature change for different $Re < 1$ and Gr, shown that ΔP_t may be approximated by the following relation:

$$\Delta P_t = \beta\frac{Gr}{Re^2\theta f(\theta)}$$

the fluctuation $f(\theta)$ is defined below in equation (6). The value of β, obtained numerically, varies between

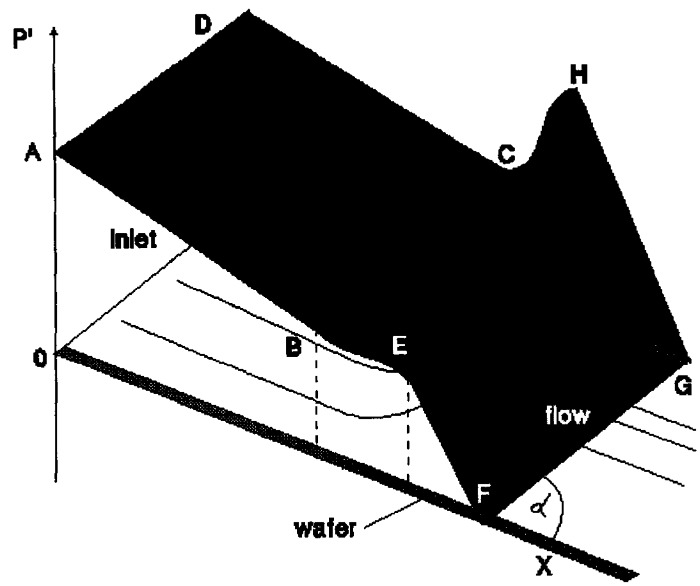


Fig. 1. $P'(x, z)$ surface. In two regions placed far upstream and downstream of transition one, the surface is coloured grey. Transition region is patterned.

0.6 and 0.7 (it rises with rising θ). Integrating equation (2), supposing linear dependence of the temperature on Z coordinate in the downstream region (it is true for the sufficiently low Re, i.e. for a weak forced convection), one can obtain the pressure change between the top and the bottom walls:

$$P_H - P_E = \frac{Gr}{Re^2 \theta f(\theta)} \tag{5}$$

where:

$$f(\theta) = \frac{\theta}{\theta - \ln(1+\theta)} \tag{6}$$

Simple geometrical consideration with applying equations (4)–(6) yields the desired condition of the return flow formation:

$$\frac{Gr}{Re} > \frac{12 L_t \theta f(\theta)}{(1-\beta)} \tag{7}$$

This formula is valid for arbitrary temperature change in the reactor. Condition (7) with a constant right hand side has been previously obtained in experiments [2] and also, intuitively by the authors of Ref. [1] (in Ref. [1] function (6) also arises in speculations, but it is not present in the final statement.)

In the case of high Reynolds numbers L_t becomes dependent on Re (directly proportion to it [4]), so that the recirculation onset depends not on the Gr/Re ratio, but on the Gr/Re^2 one. The latter relation has a clear physical meaning: at high Reynolds numbers transition to the vortical flow is defined by the ratio of the gravitational force to the inertial one (viscose force has no influence on the process). At low Re numbers viscose force has a significant impact on the flow structure, and relation Gr/Re reflects this fact, since it is the ratio of the gravitational force to the viscose force.

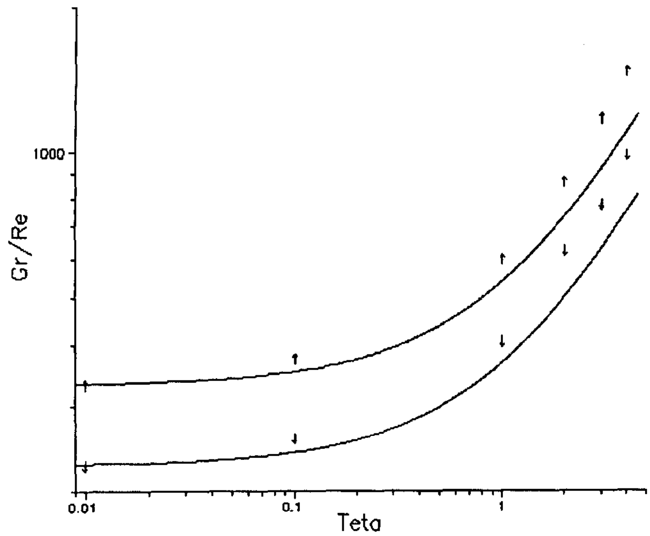
TEMPERATURE DEPENDENCE OF THE RETURN FLOWS ONSET CONDITION

It seems to be quite natural, that at least qualitative temperature dependence of the return flow onset condition has been derived in the form of equation (7): the greater is the temperature change in the reactor, the less is the P_H/P_E at fixed Gr/Re^2 relation, as it follows from equation (5), and therefore, at greater Gr/Re^2 ratio the positive horizontal component of P' gradient arises (ΔP_t diminishes slower than $P_H - P_E$ because of $\beta < 1$).

To check this conclusion, the series of calculation has been performed. The dependence of the critical relation Gr/Re^n (where $n = 1$ for $Re < 1$, and $n = 2$ for $Re \gg 1$) on θ is represented in [Fig. 2(a)]. By solid curves in the same figure the Gr/Re^n dependence given by equation (7) and coinciding with the results of calculations in the point $\theta = 0.01$ is represented for comparison. At least qualitative correspondence occurs. Quantitative coincidence may be easily obtained supposing the linear dependence $\beta = A\theta + B$, as it is shown in [Fig. 2(b)], where coefficients A and B are constant for any θ. To achieve better correspondence one can suppose A and B to be piecewise constant in two regions: $0 < \theta < 1$ and $1 \leqslant \theta$.

Some additional considerations may be proposed to clarify the obtained results. The energy conservation equation, with the temperature as an independent variable, is the homogeneous one (all its terms contain the temperature to power one). At sufficiently low Re, this equation becomes independent of the velocity field. Therefore, as θ changes, the equation solution is simply scaled without changing its shape. In other words, $T'(x, z)/\theta$ is independent on θ.

Let us write down now the Navier–Stokes equations (U and V are the X and Z components of the velocity,



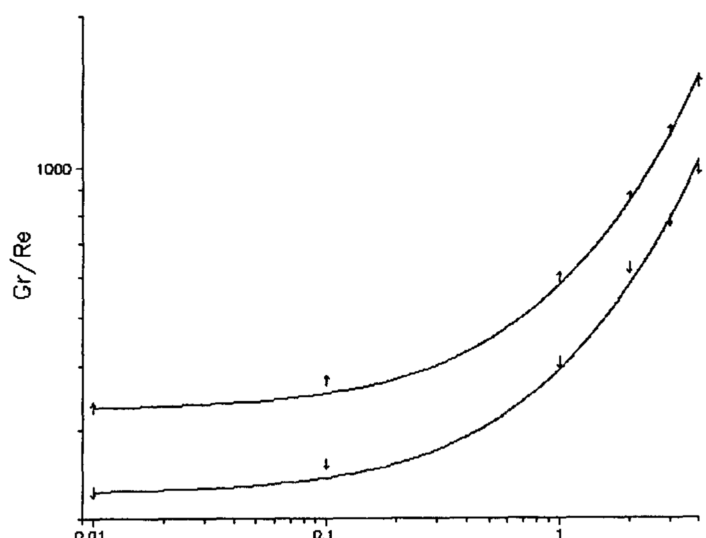


Fig. 2. The critical relation Gr/Re ($Re < 1$) and $10{*}Gr/Re^2$ ($Re > 1$) dependence on the relative temperature change in the reactor: by arrows facing down the calculated points for low Re are marked, by the up-facing ones the same is done for $Re > 1$. Solid lines show the function, given by equation (7), which coincides with the calculated data in the point $\theta = 0.01$: (a) β in equation (7) is constant; (b) β varies linearly with θ.

respectively):

$$\rho U\frac{\partial U}{\partial x}+\rho V\frac{\partial U}{\partial z}=\frac{1}{Re}\left(\frac{4}{3}\frac{\partial^2 U}{\partial x^2}+\frac{\partial^2 U}{\partial z^2}+\frac{1}{3}\frac{\partial^2 V}{\partial z\,\partial x}\right)-\frac{\partial P'}{\partial x}; \tag{8}$$

$$\rho U\frac{\partial V}{\partial x}+\rho V\frac{\partial V}{\partial z}=\frac{1}{Re}\left(\frac{4}{3}\frac{\partial^2 V}{\partial z^2}+\frac{\partial^2 V}{\partial x^2}+\frac{1}{3}\frac{\partial^2 U}{\partial x\,\partial z}\right)-\frac{\partial P'}{\partial y}+\frac{Gr}{Re^2\theta}\frac{T'}{1+T'}; \tag{9}$$

and note, that the last term in the right-hand side of equation (9) is the only one where the temperature field appears in these equations. When $\theta \ll 1$, this term may be written as $(Gr{*}Re^2)(T'/\theta)$ and its independent of θ. Therefore, the velocity field does not depend on θ and ΔP_t has some constant value ΔP_{t0}.

When the temperature change in the reactor rises, the source term in the right hand side of equation (9) decreases because T' increases the denominator. Therefore, the Navier–Stokes equations tend to their isothermal form for high θ, and that is why ΔP_t drops as the temperature change in the reactor increases.

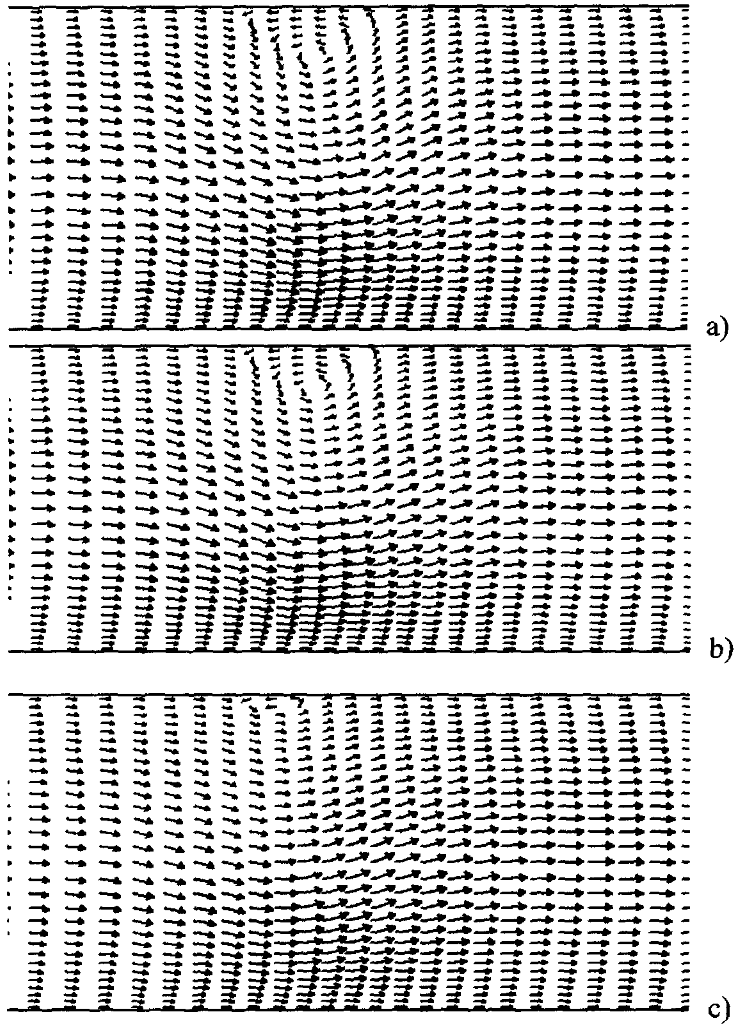


Fig. 3. The vortex flow in the horizontal reactor at different hot wall temperatures. The same reactor region is depicted for $Re = 0.1$, $Gr = 30$: (a) $\theta = 0.01$; (b) $\theta = 0.1$; (c) $\theta = 1.0$.

Several interesting facts, that qualitatively supplement speculations presented above, have been noted while performing calculations. First, with given Gr/Re relation (at low Re) positive horizontal gradient of P' is either present for all investigated θ or is not present at all. If it existed for some θ, then with rising temperature of the hot wall the absolute positive change of P' (between the local minimum and maximum of P' on the top wall, see Fig. 3) diminished, but never vanished. If for some small θ_0 vortex occurred, then for $\theta_0 < \theta \ll 1$, the flow structure changed slightly, while for θ of the order or greater than unity, the center of the vortex moved towards the top wall, the vortex, being 'compressed' by expanding gas, became weaker (it is demonstrated in Fig. 3) and at some value of θ vanished. Sometimes it was possible to 'find' the lost vortex by refining the grid properly (in other words, by resolving the vortex on the grid). On the other hand, if there was not positive horizontal P' gradient at some value of the temperature change, it never observed at any θ, consequently, the vortices were not observed too. One can assume, that when the positive P' gradient presents, there is at least theoretical possibility to resolve the vortex by refining the grid. The real vortex disappearance takes place when the vortex size becomes of the order of the gas fluctuations size.

THREE-DIMENSIONAL EFFECTS, ACCOMPANYING THE VORTEX FORMATION IN HORIZONTAL CVD REACTOR

It is not possible to investigate the vortex influence on the flow symmetry by means of the two-dimensional model. Nevertheless, this question is especially

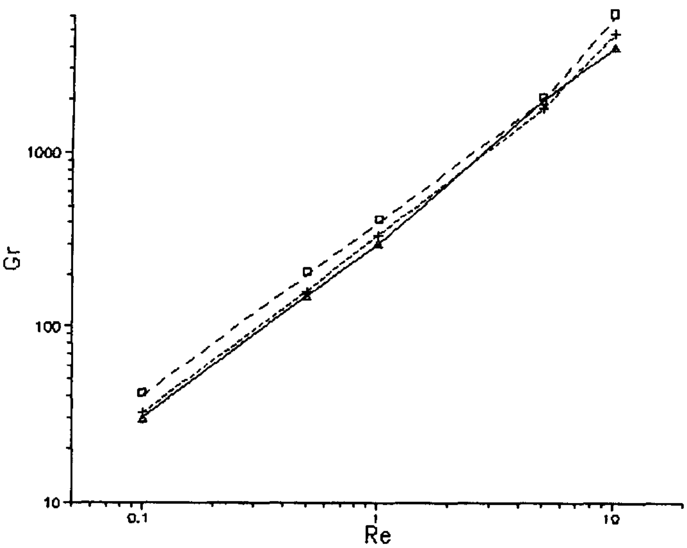


Fig. 4. Gr_{cr} (minimum Gr such that for all $Gr > Gr_{cr}$ recirculation presents) dependence on Re in a wide reactor: points, marked by the triangles and linked by the solid line are obtained for the 3D reactor with insulating side walls, the dashed line and the squares mark Gr_{cr} in the case of cooled side walls, the dotted line and the crosses denote results for the 2D model.

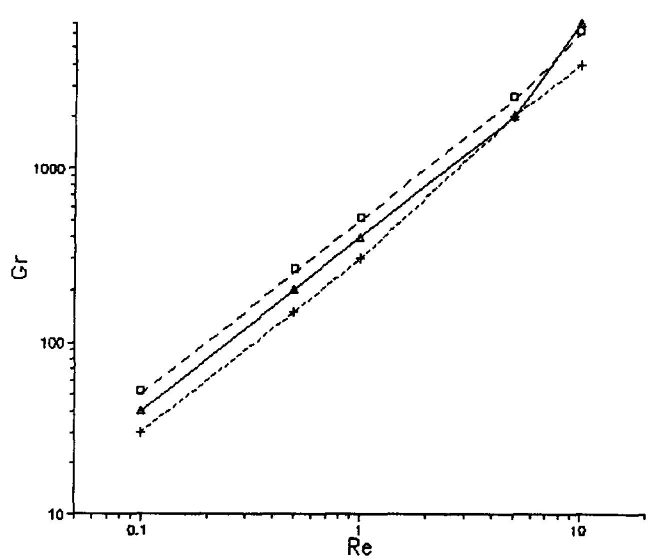


Fig. 5. Gr_{cr} dependence on Re in the narrow 3D reactor. The symbols' meaning is the same as in Fig. 4, except for the dotted line and the crosses, that show Gr_{cr} for the wide 3D reactor with the insulating walls.

important for the 2D model performance, since the symmetry violation makes the 2D model inapplicable. To study the possibility of applying the 2D model to simulate vortical flows, the 3D model program realisation has been implemented and the series of calculations has been performed. The parallel piped-shaped reactor was considered. The gas inlet was placed on one of the reactor's vertical walls, the outlet was located on the opposite one. The heated wafer was placed on the bottom wall. The boundary conditions on the side and top walls were prescribed either isothermal or adiabatic. The applied solution method was the same, as for the 2D case. All the calculations were made for $\theta = 2$.

Some results of calculations for different types of reactor (relation length $\times$ height $\times$ width was $15 \times 1 \times 5$ for the wide reactor and $15 \times 1 \times 2$ for the narrow one) and for different kinds of boundary condition are represented in Figs. 4 and 5. Several conclusions may be made using the results of calculations:

(1) Recirculation always arises near the side walls. It is always less developed in the reactor centre, and at the very beginning of the vortex formation process

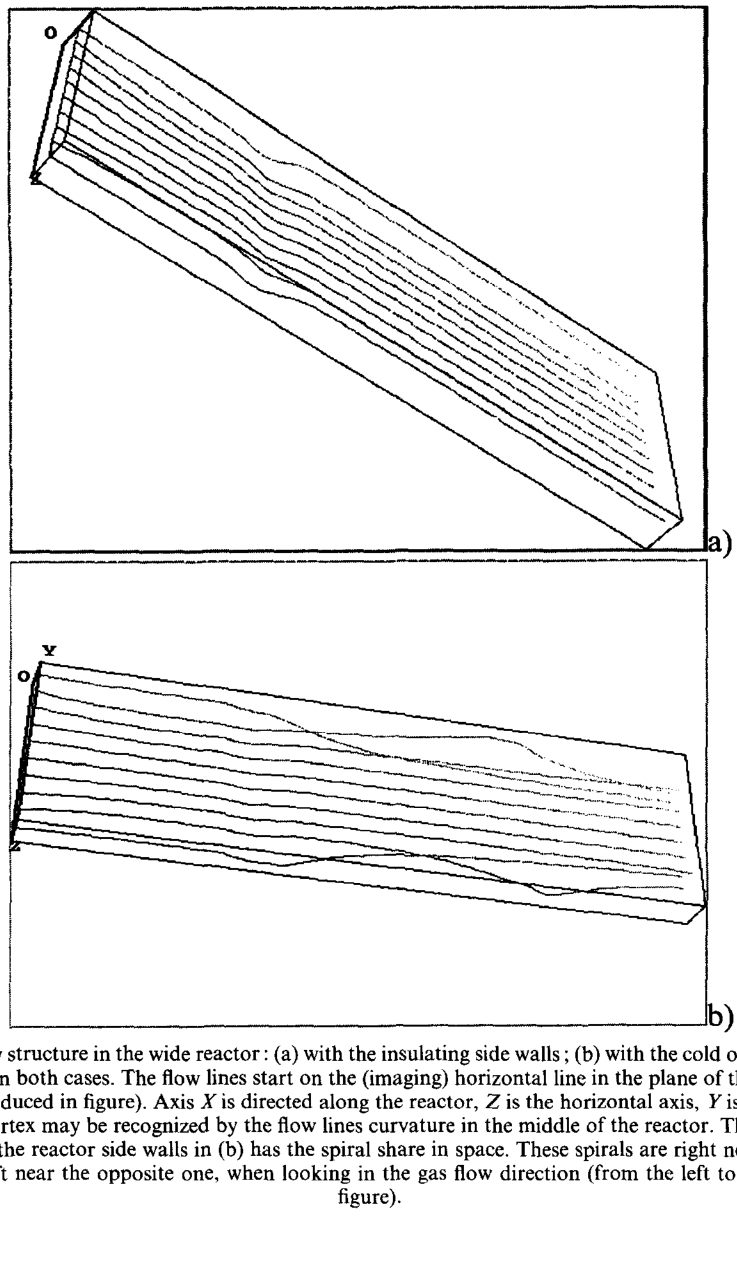


Fig. 6. Flow structure in the wide reactor: (a) with the insulating side walls; (b) with the cold ones. $Re = 5$, $Gr = 2000$ in both cases. The flow lines start on the (imaging) horizontal line in the plane of the inlet (this line is reproduced in figure). Axis X is directed along the reactor, Z is the horizontal axis, Y is the vertical one. The vortex may be recognized by the flow lines curvature in the middle of the reactor. The wave-like lines along the reactor side walls in (b) has the spiral share in space. These spirals are right near the right wall and left near the opposite one, when looking in the gas flow direction (from the left to the right in figure).

(low Gr/Re) there may be no recirculation at all in the bulk gas (while near the walls the recirculation presents). A conclusion may be made, that the 2D model solution, which is most reliable near the reactor mid-plane, gives the values of critical Gr greater than they really are. It is confirmed by the 2D and 3D results comparison, see Fig. 4. Anyway, the difference between the 2D and 3D models' predictions is sufficiently small and is explained by the boundary effects near the side walls. The return flow itself does not cause the 2D symmetry break.

(2) The cooled walls hinder the vortex formation comparatively to the adiabatic ones. This fact may be caused by the longitudinal rolls formation. The nature of these rolls is similar to that of the Rayleigh–Benard convection [5]. The well-known fact is that in the reactor with cooled walls this phenomena takes place at all non-zero Gr. The longitudinal rolls arise near the side walls, and the region which they occupy rises with rising Gr. In Fig. 6 the gas flow in the wide reactor is represented for the two different thermal regimes on the side walls (all other things being the same). The spiral structures are clearly recognised near the cooled walls [Fig. 6(b)]. With rising Grashof number these structures occupy the entire reactor volume. It indicates an appreciable transport process in the lateral direction, and, consequently, shows the 2D model to be inadequate. In the narrow reactor the same process goes at lower Gr, and the 2D model is even less inadequate than it may be assumed, say, on the basis of the boundary effects analysis in the isothermal Poiseuille flow. Hence the best accuracy of the 2D model is

achieved for the wide reactor with heat-insulating walls (it has to be noted, that the longitudinal rolls in the reactor with the insulating walls arise anyway, but only for high Gr, starting approximately from $Gr \sim 2440$ [5]). Reactor with the cold walls, and especially the narrow reactor are described by the 2D model adequately only at sufficiently low Gr (for instance, Ref. [6]).

(3) Recirculation in the narrow reactor takes place at higher Gr, than in the wide reactor.

(4) The power type of critical relation Gr/Re^n remains unchanged regardless of the boundary conditions type prescribed on the side walls and of the reactor width, at least, at low Re.

CONCLUSIONS

The approach proposed in section 1 of the present work offers clear explanation for the return flow formation mechanism in a horizontal reactor with the one wall being heated and the opposite one being cooled. The recirculation onset condition (7) coincides in form with the empirical one and is confirmed by the results of numerical investigation. It can easily be expanded to the case of high Reynolds numbers, and allows to explain the temperature dependence of the recirculation onset condition.

Three-dimensional modeling has shown, that the cooled side walls, as well as a small width of the reactor suppress the return flow formation (as compared to the wide reactor with insulating side walls). The interesting fact is that for $Re < 1$ the recirculation flow onset condition conserves its form regardless of the type of flow: 2D or 3D.

Acknowledgements—Fruitful support by K. A. Valiev and A. V. Panyukhin is gratefully acknowledged.

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APPENDIX: THE GAS FLOW GOVERNING EQUATIONS AND THE METHOD OF THEIR SOLUTION

The low-Mach number approximation of the gas dynamics equations has been used:

$$\frac{\partial \rho}{\partial t} + \nabla(\rho \mathbf{V}) = 0 \tag{A1}$$

$$\frac{\partial \rho \mathbf{V}}{\partial t} + \nabla(\rho \mathbf{V}\mathbf{V}) = \frac{1}{Re}\nabla(\mu(\mathbf{V}+\mathbf{V}^{+}) - \frac{2}{3}\mu(\nabla\mathbf{V})\mathbf{I}) - \nabla P' + \frac{1}{Fr}\frac{\mathbf{g}}{g_0}(\rho-\rho_0) \tag{A2}$$

$$C_p\left(\frac{\partial \rho T'}{\partial t} + \nabla(\rho \mathbf{V} T')\right) = \frac{1}{Re\,Pr}\nabla(\lambda \nabla T') \tag{A3}$$

These equations were discretized by means of the finite differences (control volume) method and solved consecutively. Coupling between the Navier–Stokes (A2) and continuity (A1) equations has been performed using the method quite similar to that used in MAC algorithm. The temperature dependence of kinetic coefficients has been approximated by the polynomials (nitrogen has been chosen as a carrier). The steady solutions have been obtained by integrating the equations in time, until the normalized residuals drop below some specified small value.

The Poiseuille velocity profile has been used as an inlet boundary condition. The flow direction has coincided with the horizontal axis direction. The vortex (return flow) was assumed to present if the negative values of the horizontal velocity component arise in a steady flow.