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Macroscopic convective phenomena in non-uniformly heated liquid mixtures

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Chronicle of one PhD dissertation on thermal convection

PhD thesis:

Glukhov Alexander Experimental Investigation of Thermal Convection in Conditions of Gravity Stratification // Perm State University, Perm, 1995. – 140 p.

Alexander F. Glukhov, 2007

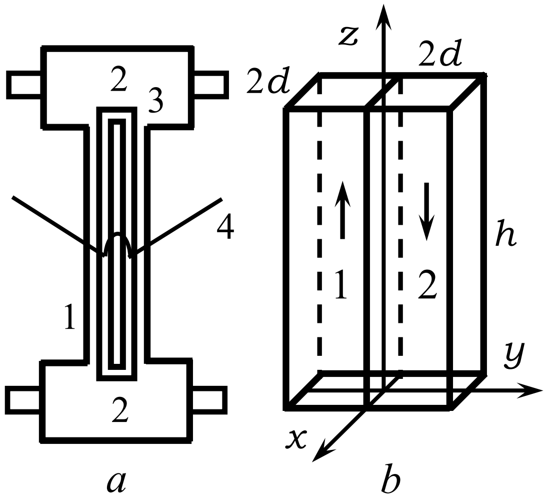
Experimental investigation had three basic parts:

- Thermal convection of liquid molecular mixtures in connected channels;
- Thermal convection of ferrofluid in connected channels;
- Evolution of particles distribution in vertical pipe filled by ferrofluid.

Our present-day evaluation of dissertation results:

- ☺ Correct paradigm was formed for concerned phenomena in molecular mixtures;
- ☺ It was obvious that thermodiffusion and sedimentation must play definite role in these phenomena;
- ☹ The contribution of each factor wasn't clear;
- ☹ Numerical model wasn't built.

Experimental data



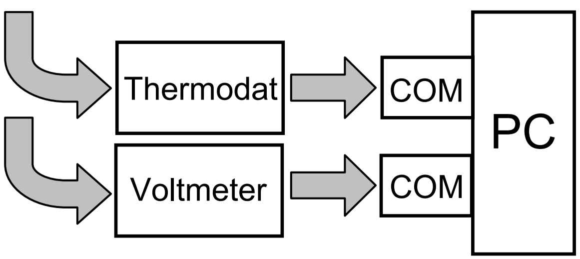


Fig. 1. Experimental setup (*a*): copper bar (*1*), heat exchangers (*2*), channels (*3*), thermocouples (*4*); coordinate system (*b*).

Width and height of the channels:

d = 3.2 mm; H = 50 mm

Binary mixtures

One of the working fluids was a mixture of Carbon Tetrachloride CCl_4 and Decane $C_{10}H_{22}$

1) Decane $C_{10}H_{22}$ (Pr = 15), 2) CCl_4 (heavy admixture)

Thermodiffusion properties of this mixture are not investigated in details until now;

Schmidt number Sc = ν/D > 1000, ε – ? (1992);

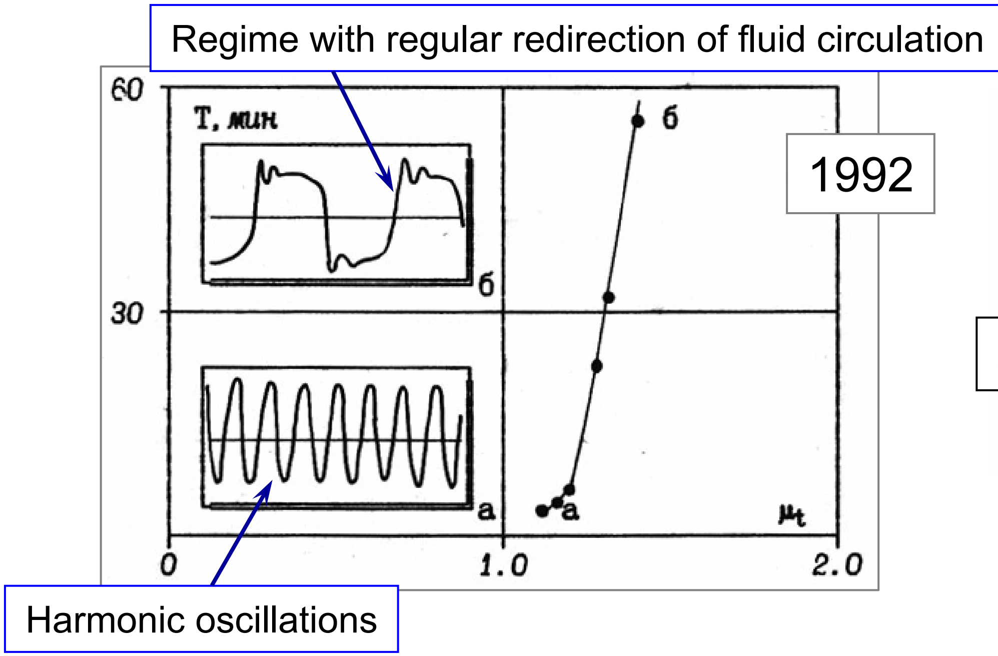
The second liquid was a mixture of water and sulphate of natrium:

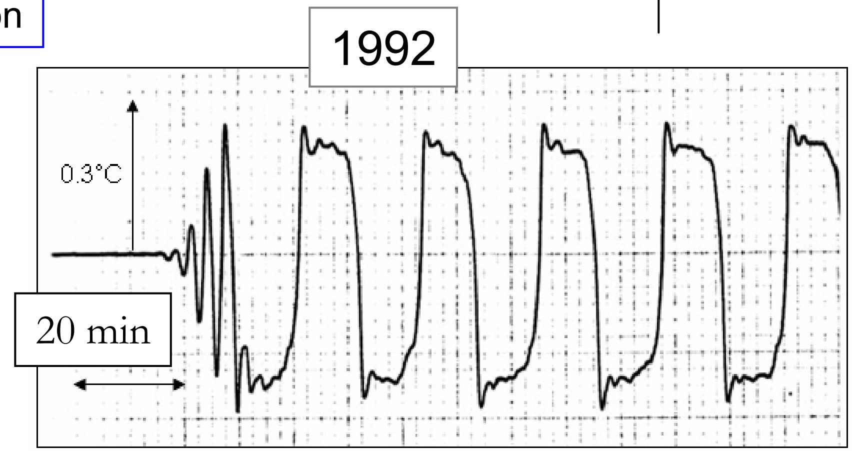
1) water H_2O (Pr = 7), 2) natrium sulphate Na_2SO_4 (heavy admixture in water)

Thermodiffusion properties of this mixture are well known:

Schmidt number Sc = 2100, parameter of thermodiffusion ε = 0.36 (2005).

Harmonic oscillations and regime with regular redirection of fluid circulation





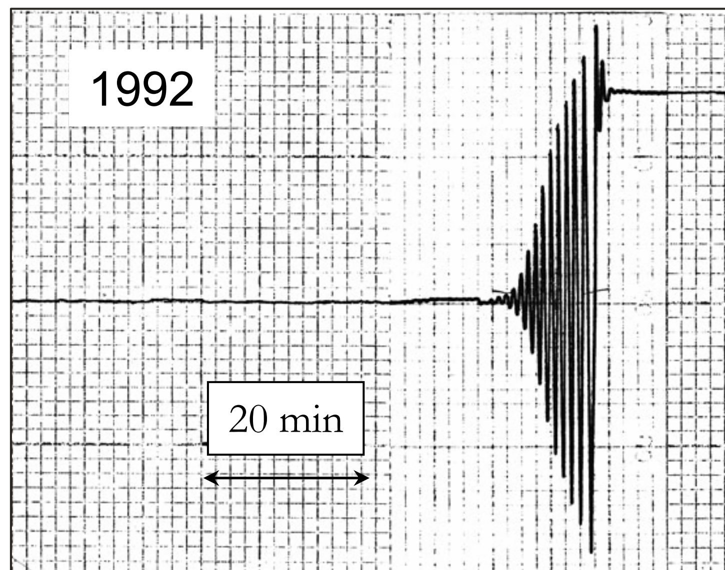


Fig. 2-4. First experimental data correspond to 5 - 15% solution of the denser CCl_4 in the less dense $C_{10}H_{22}$.

The shape of the oscillations was transformed from near-sinusoidal to near-rectangular with the growth of supercriticality.

Principal explanation for molecular mixtures

Mechanism responsible for the effects observed in experiments is mainly attributable to the thermodiffusion separation of the mixture which is due to the horizontal temperature gradients $\theta/d = 3$ K/cm rather than to the weak vertical gradients $\Theta/h = 0.3$ K/cm with a characteristic component separation time h^2/D ~ 103 hours; h – height, d – width of the channel.

Horizontal gradients occur only in the circulating fluid. The separation time across the channel is d^2/D ~ 1 hour, which coincides in order of magnitude with the time of circulation of the fluid around the loop.

Liquid particle changes itself composition during the motion in each channel.

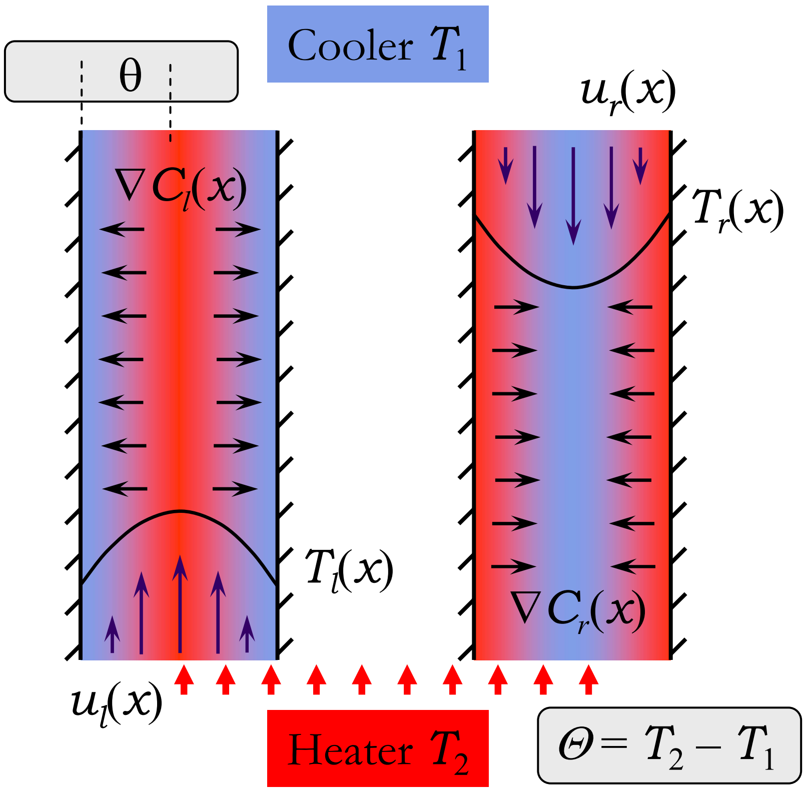
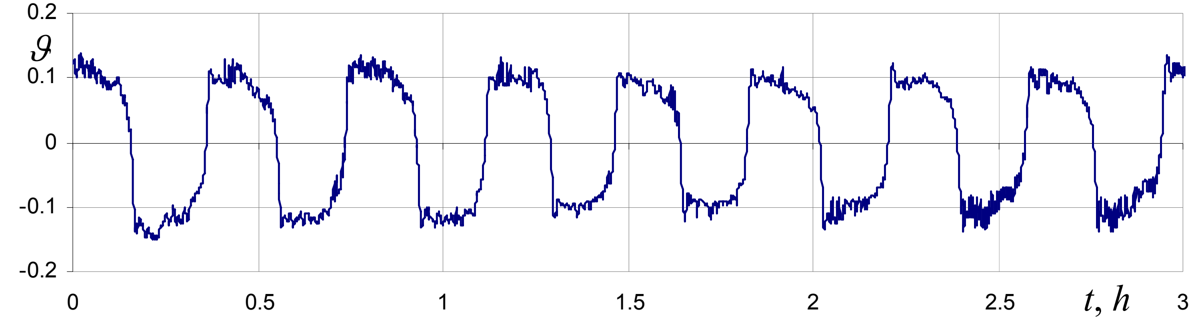


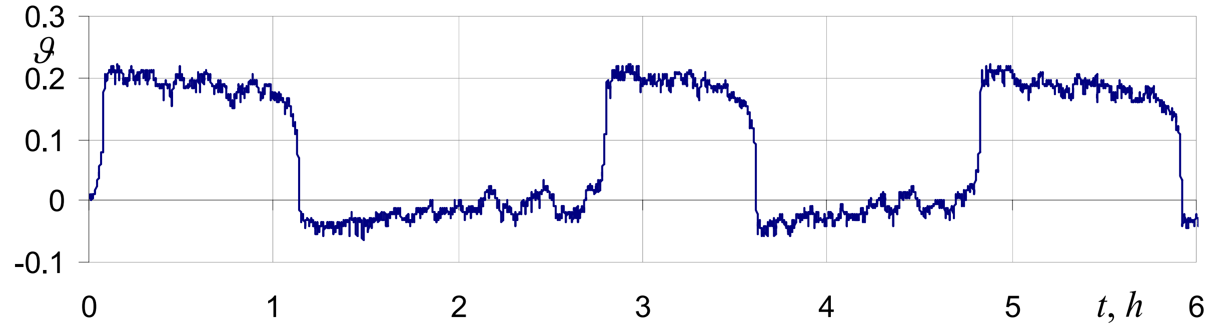
Fig. 5. Schematic visualization of the admixture distribution. Left channel accumulates heavy component, right one loses it.

Flow of ferrofluid with regular redirection of circulation

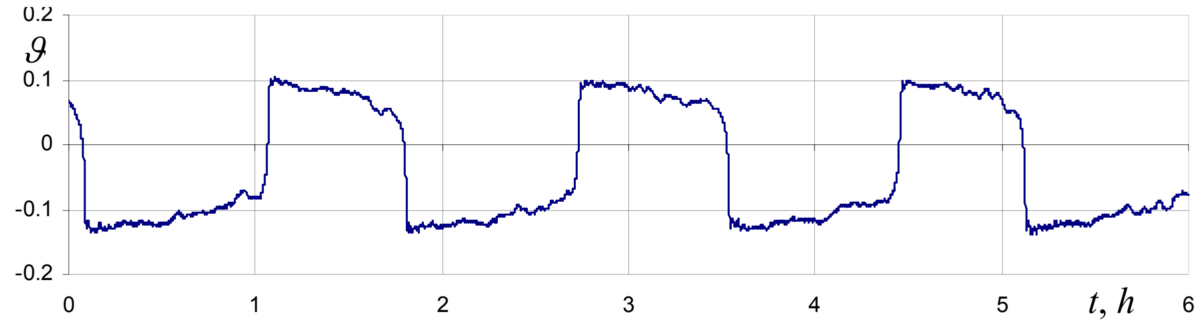
Solution of Na_2SO_4 in water, 16%;
$\Theta = 10$ °C,



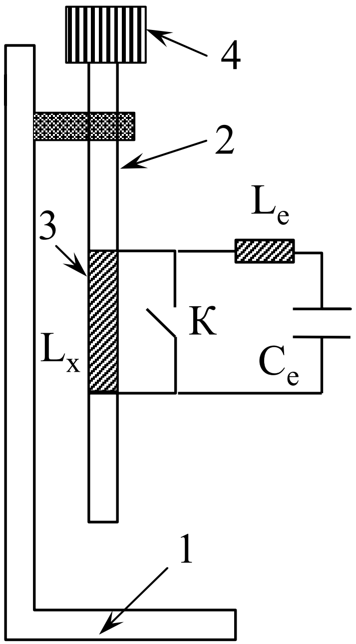
Ferrofluid, 4%;
$\Theta = 2.0$ °C,
$\nabla\phi = 0.58 \cdot 10^{-5}$ cm^{-1}
$f = 0.56 \cdot 10^{-2}$ c^{-1}

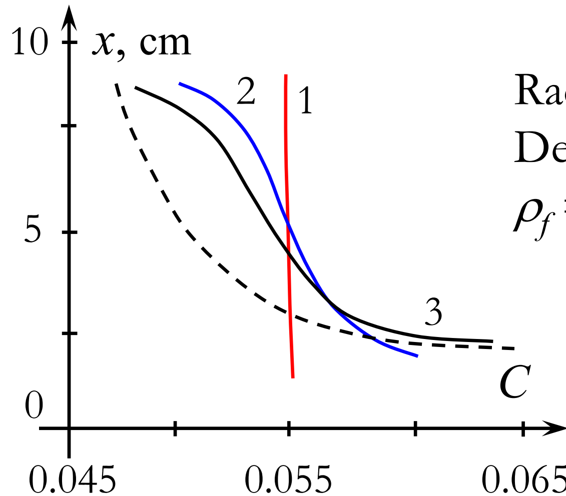


Ferrofluid, 12%;
$\Theta = 6.0$ °C,
$\nabla\phi = 2.62 \cdot 10^{-5}$ cm^{-1}
$f = 0.72 \cdot 10^{-2}$ c^{-1}



Sedimentation in ferrofluid





Radius of particle: 10 nm;
Density of kerosene and magnetite:
$\rho_f = 0.82\ \text{g/cm}^3$; $\rho_m = 5.5\ \text{g/cm}^3$

1 – 40 hours
2 – 6300 hours
3 – 10300 hours

Fig. 6. Scheme of experimental setup for the measurement of magnetic particles concentration and concentration in dependence on height for different moment of time.

1 – metal support; 2 – test-tube with ferrofluid on the base of kerosene; 3 – inductive sensor of particles concentration; 4 – screw to move the test-tube

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Conclusion: The effect of particles sedimentation exists and can be estimated even in the beginning of thermal convection with the help of the formula for frequency of transitional oscillations :

$$\nabla\phi = \frac{\beta_t}{\beta_\phi}\frac{d^4 \nabla T}{\pi^2 \chi^2} f^2 = 10^{-4} \div 10^{-5}\ \text{cm}^{-1}$$

f – frequency of transitional oscillations

Does convective behaviour of molecular mixtures and ferrofluids have common nature or only individual common features?

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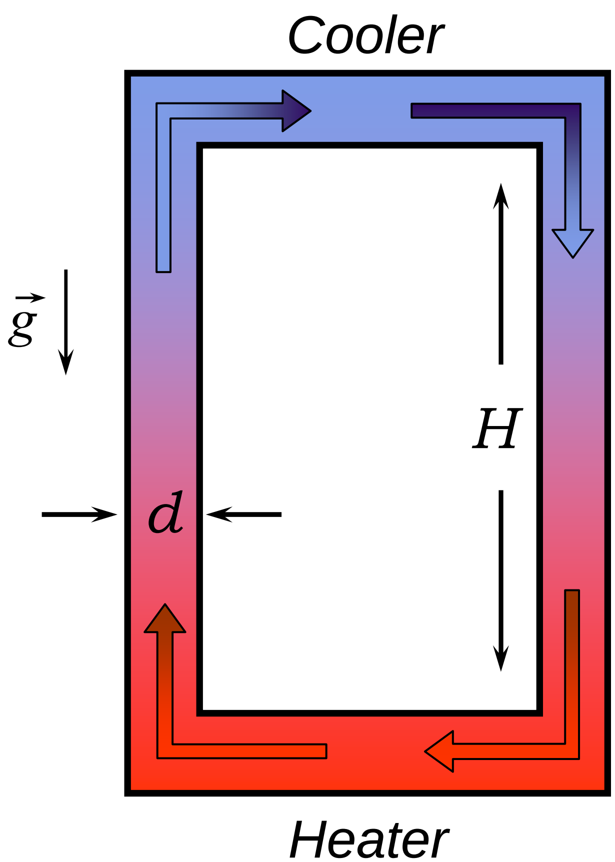
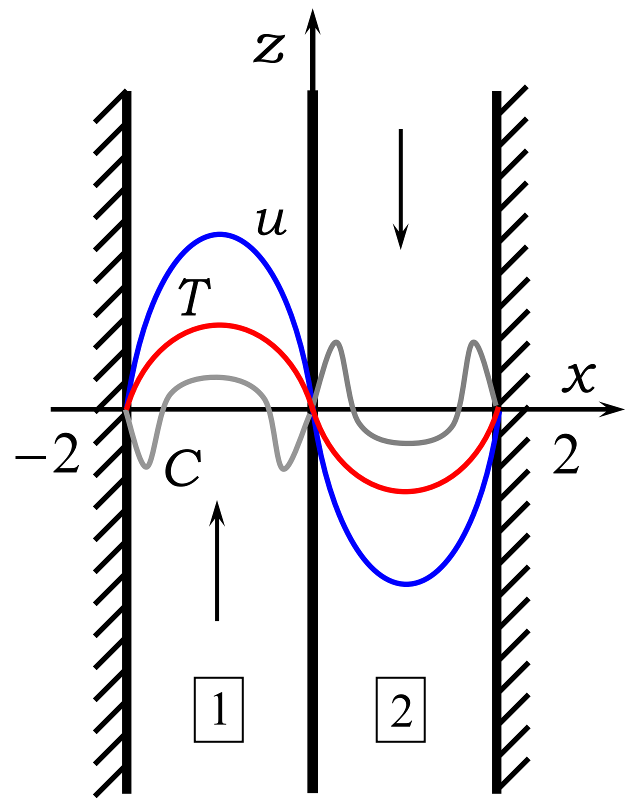
How can elephant be eaten?

Answer:

Only bit by bit.

Binary molecular mixtures





Physical requirement: $H >> d$

1) Straight-trajectories approximation is applied **2)** Boundaries of channels have high heat conductivity **3)** Antisymmetric solutions for fields of temperature, velocity and concentration are valid.

Basic assumptions

The diffusion and heat fluxes are related with the concentration and temperature gradients in general case by the formulas:

$$\vec{j} = -\rho D\left(\nabla C + \alpha \nabla T\right) \qquad \vec{q} = -\left(\lambda + \alpha D \Lambda\right)\nabla T - D\Lambda \nabla C$$

The effects associated with the presence of an admixture are characterized by the coefficients of diffusion D and thermodiffusion α.

Expansion of density:

$$\rho = \rho_o\left(1 - \beta_t T + \beta_c C\right)$$

The concentration density coefficient β_c describes the dependence of the density on the concentration:

$$\beta_c = \frac{1}{\rho_o}\left(\frac{\partial \rho}{\partial C}\right)_{T,p}$$

The equations for an incompressible fluid in the Boussinesq approximation had been used to simulate the convective flows of a binary mixture:

$$\frac{\partial \vec{v}}{\partial t} + (\vec{v}\nabla)\vec{v} = -\frac{1}{\rho_o}\nabla p + \nu \Delta \vec{v} + g\left(\beta_t T - \beta_c C\right)\vec{\gamma} \qquad \frac{\partial C}{\partial t} + (\vec{v}\nabla)C = D\Delta C + \alpha D \Delta T$$

$$\frac{\partial T}{\partial t} + (\vec{v}\nabla)T = \left(\chi + \alpha^2 D\Lambda\right)\Delta T + \alpha D \Lambda \Delta C$$

C – mass concentration of heavy admixture

Shaposhnikov I.G. Theory of Convective Phenomena in a Binary Mixture // Prikl. Matem. Mekh., 1953.

Equations in non-dimensional form and control parameters

Units:

- Length $L - [2d]$,
- Velocity $v - [\nu/d]$,
- Time $t - [d^2/\nu]$,
- Pressure $p - [\rho_o \nu^2/d^2]$,
- Concentration $C - [\Theta\beta_t/\beta_c]$,
- Temperature $T - [\Theta]$.

Here Θ is the temperature difference between heat exchangers.

$$\frac{\partial \vec{v}}{\partial t} + \frac{1}{Pr}(\vec{v}\nabla)\vec{v} = -\nabla p + \Delta \vec{v} + \frac{RaH}{Pr}(T - C)\vec{\gamma}, \qquad \mathrm{div}\,\vec{v} = 0,$$

$$\frac{\partial T}{\partial t} + (\vec{v}\nabla)T = \frac{1}{Pr}\Delta T, \qquad \frac{\partial C}{\partial t} + (\vec{v}\nabla)C = \frac{1}{Sc}(\Delta C + \varepsilon \Delta T).$$

Nondimensional parameters: thermal Rayleigh number, thermodiffusive parameter, Prandtl and Schmidt numbers:

$$Ra = \frac{g\beta_t d^4}{\nu\chi}\frac{\Theta}{h}$$

$$\varepsilon = \frac{\alpha\beta_c}{\beta_t}$$

$$Pr = \frac{\nu}{\chi}$$

$$Sc = \frac{\nu}{D}$$

Boundary conditions on vertical walls for field of concentration:

$$\left.\frac{\partial C}{\partial \vec{n}}\right|_\Gamma + \varepsilon \left.\frac{\partial T}{\partial \vec{n}}\right|_\Gamma = 0.$$

Mechanical equilibrium state

Conditions of mechanical equilibrium:

$$\vec{v}=0,\ p=p_o,\ T=T_o,\ C=C_o,\ \frac{\partial}{\partial t}=0,$$

Boundary condition on concentration:

$$\left.\frac{\partial C}{\partial \vec{n}}\right|_\Gamma + \varepsilon \left.\frac{\partial T}{\partial \vec{n}}\right|_\Gamma = 0.$$

Equations system:

$$\Delta T_o = 0, \quad \Delta C_o + \varepsilon \Delta T_o = 0, \quad (\nabla T_o - \nabla C_o)\times \vec{\gamma} = 0$$

Equilibrium distributions of temperature and concentration:

$$\begin{cases} T_o(z) = -z/H, \\ C_o(z) = \varepsilon z/H. \end{cases}$$

Stationary flow:

$$\vec{v}(\ 0,\ 0,\ U(x,y,t)\ )$$

It's possible to use the straight - trajectory approximation.

Fields distributions in cross-section:

$$F(x,y,z) = f(z)\mathrm{s}_{13}(x)\mathrm{c}_{13}(y) \qquad F = C + \varepsilon T$$

$$U(x,y) = u\cdot \sin\left(\frac{\pi x}{2}\right)\cos\left(\frac{\pi y}{2}\right)$$

$$T(x,y,z) = \theta(z)\sin\left(\frac{\pi x}{2}\right)\cos\left(\frac{\pi y}{2}\right)$$

$$\mathrm{s}_{13}(x) = \sin\left(\frac{\pi x}{2}\right) - \frac{1}{3}\sin\left(\frac{3\pi x}{2}\right)$$

$$\mathrm{c}_{13}(y) = \cos\left(\frac{\pi y}{2}\right) + \frac{1}{3}\cos\left(\frac{3\pi y}{2}\right)$$

Stationary solution

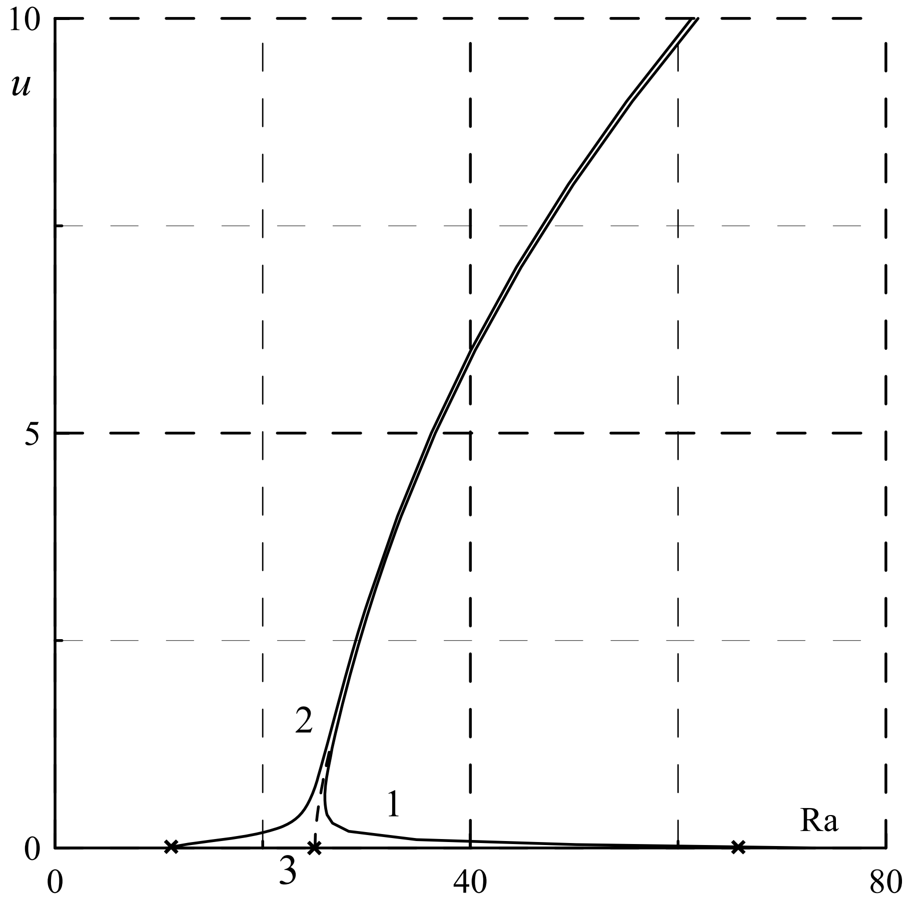


Fig. 7. Amplitude curves for different values of thermodiffusion parameter:

1 – ε = – 0.01; 2 – ε = 0.02; 3 – ε = 0.

There is formula in limit $u \to 0,\ \varepsilon = 0$:

$$\mathrm{Ra}_c = \frac{\pi^4}{4\left(1 - \dfrac{1}{z_1}\tanh z_1\right)} \qquad z_1 = \pi H / 2\sqrt{2}$$

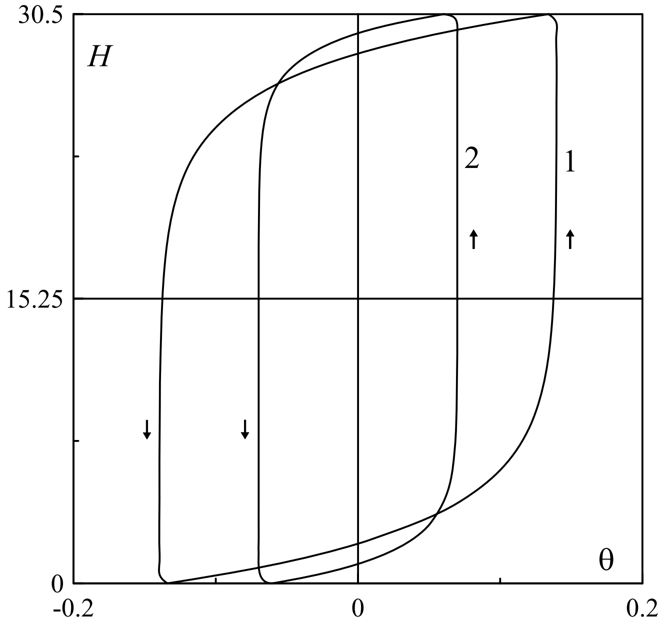
In limiting case $H \to \infty$ formula gives well-known value of critical Rayleigh number $\mathrm{Ra} = \pi^4/4$.

Solution for arbitrary values of thermodiffusion parameter, Prandtl and Schmidt numbers:

$$\mathrm{Ra}_c = \frac{\pi^4}{4}\left\{(1+\varepsilon)\left(1 - \frac{1}{z_1}\tanh z_1\right) + \frac{\varepsilon \mathrm{Sc}}{\mathrm{Pr}}\left(0.45 - \frac{1}{z_2}\tanh z_2\right)\right\}^{-1} \qquad z_2 = \frac{3\sqrt{10}\pi H}{20}$$

Non-stationary regimes

$\vec{v}(\ 0,\ 0,\ u(x,y,t)\ )$ – Straight-trajectory approximation is valid as before.



Fourier analysis of experimental data indicates that fields distributions can be approximated by several trigonometric functions:

$$u(x,y,t), \qquad T = T_1(x,y,t)\sin\left(\frac{\pi z}{H}\right) + T_2(x,y,t)\cos\left(\frac{\pi z}{H}\right)$$

$$F = F_1(x,y,t) + F_2(x,y,t)\cos\left(\frac{\pi z}{H}\right) + F_3(x,y,t)\cos\left(\frac{2\pi z}{H}\right)$$

Fig. 8. Temperature distribution along vertical axis. The arrows show flow direction in the channels.

The values of velocity are: 1 – u = 3; 2 – u = 1.5

Glukhov A.F., **Demin V.A.**, and **Putin G.F.** Separation of Mixtures, Heat and Mass Transfer in Connected Channels // Technical Physics Letters, Vol. 34, No. 9 (2008).

Equations system for amplitudes

$$\begin{cases} \dfrac{\partial T_1}{\partial t} - \dfrac{\pi}{H} u T_2 = \dfrac{1}{Pr} \hat{\Pi} T_1 - \dfrac{\pi^2}{Pr H^2} T_1 + \dfrac{4}{\pi H} u, \\ \dfrac{\partial T_2}{\partial t} + \dfrac{\pi}{H} u T_1 = \dfrac{1}{Pr} \hat{\Pi} T_2 - \dfrac{\pi^2}{Pr H^2} T_2, \end{cases}$$

$$\hat{\Pi} = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$$

$$\left\{ \frac{\partial u}{\partial t} = \hat{\Pi} u + \frac{RaH}{Pr} \left(\frac{2}{\pi} T_1 (1 + \varepsilon) - F_1 \right), \right.$$

$$\begin{cases} \dfrac{\partial F_1}{\partial t} - \dfrac{2}{H} u F_2 = \dfrac{1}{Sc} \hat{\Pi} F_1 + \dfrac{2\varepsilon}{\pi Pr} \hat{\Pi} T_1 - \dfrac{2\pi\varepsilon}{H^2 Pr} T_1, \\ \dfrac{\partial F_2}{\partial t} - \dfrac{16}{3H} u F_3 = \dfrac{1}{Sc} \hat{\Pi} F_2 - \dfrac{\pi^2}{H^2 Sc} F_2 + \dfrac{\varepsilon}{Pr} \hat{\Pi} T_2 - \dfrac{\pi^2 \varepsilon}{H^2 Pr} T_2, \\ \dfrac{\partial F_3}{\partial t} + \dfrac{4}{3H} u F_2 = \dfrac{1}{Sc} \hat{\Pi} F_3 - \dfrac{4\pi^2}{H^2 Sc} F_2 + \dfrac{4\varepsilon}{3\pi Pr} \hat{\Pi} T_1 - \dfrac{4\pi\varepsilon}{3H^2 Pr} T_1 \end{cases}$$

- Equations were solved numerically with the help of a finite-difference method.
- Computer module was written using the programming language “FORTRAN-90.”
- The algorithm was designed in accordance with the explicit solution scheme.
- The calculations were executed using the time-relaxation method.

Glukhov A.F., Demin V.A., Putin G.F. // Fluid Dynamics, Vol. 42, No. 2 (2007).

Summary results for binary molecular mixtures

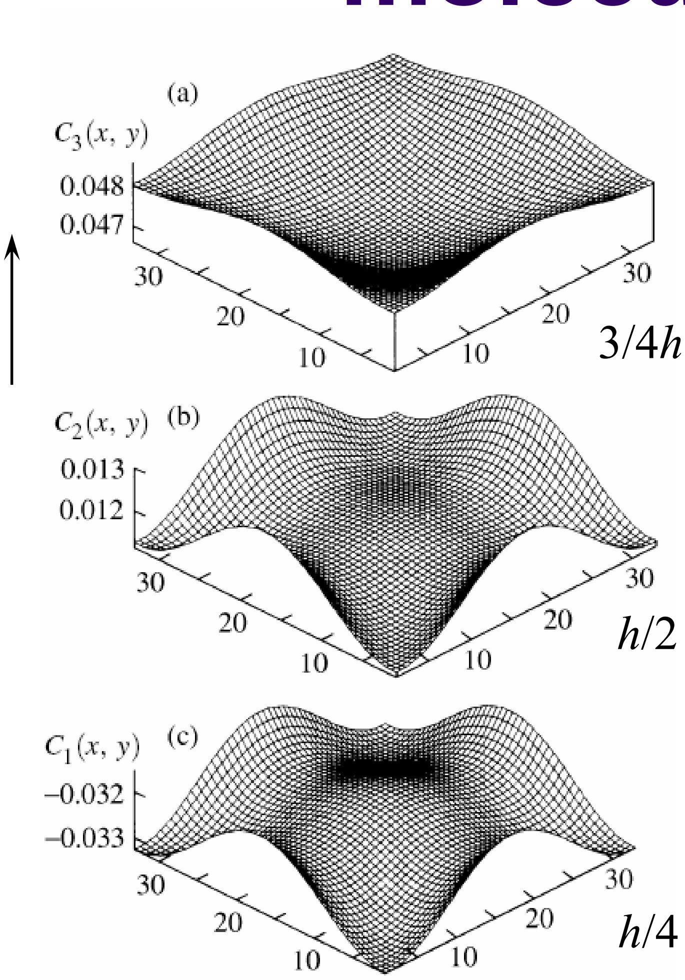
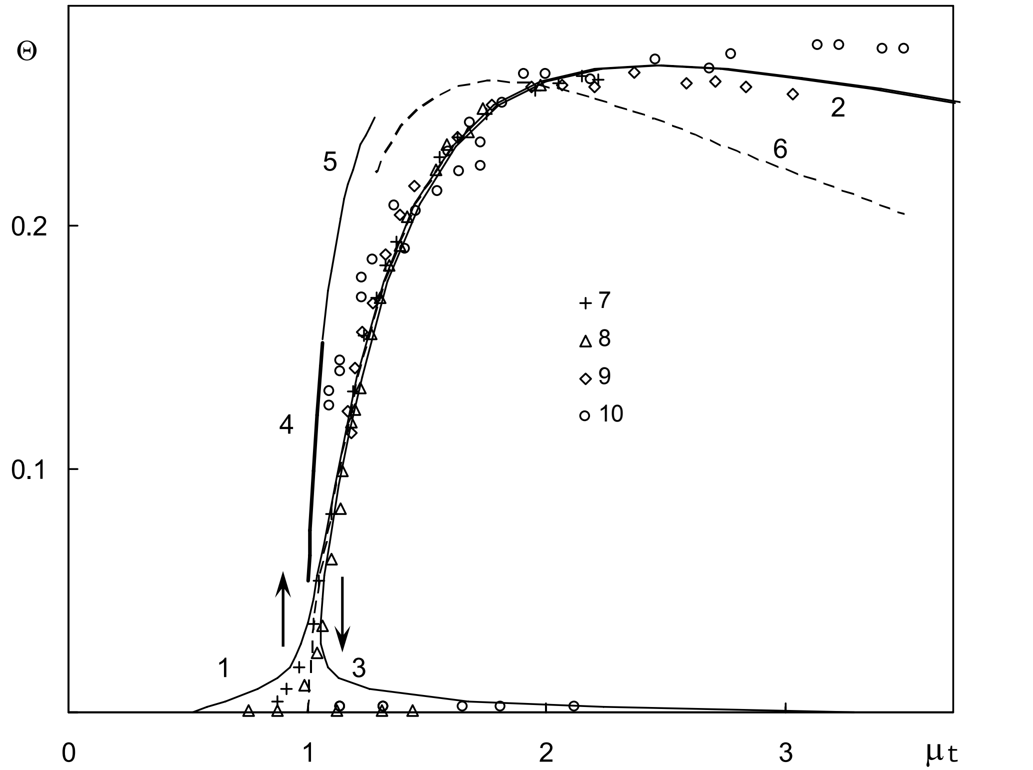


Fig. 9. Fields of admixture concentration in cross-section along channel



Amplitude curves: (*1*), (*2*), (*3*) – stationary flows for $\varepsilon > 0$, $\varepsilon = 0$, $\varepsilon < 0$ respectively; (*4*), (*5*) – amplitudes of the harmonic and “flop-over” oscillations; (*6*) – steady-state regimes for high values of supercriticality; (7-*10*) – experimental data; the arrows show the “hard” transitions from equilibrium to intense convection and the transition back to equilibrium.

Last experiments with ferrofluids and binary mixtures

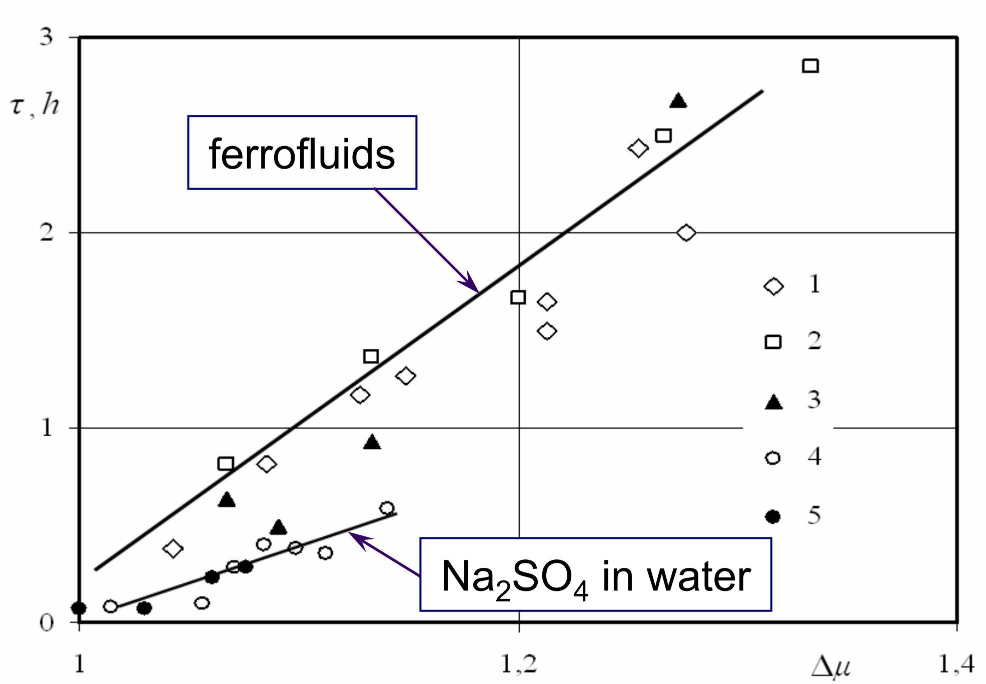


Fig. 10. Period of “flop-over” oscillations:

1 – Ferrofluid, **12%** (ΔT_c = 4.7 K);

2 – Ferrofluid, **4%** (ΔT_c = 1.5 K);

3 – Kerosene, particles concentration **0%**;

4 – Na_2SO_4 in water, **10%** (ΔT_c = 7.1 K);

5 – Na_2SO_4 in water, **4%** (ΔT_c = 6.6 K).

First line corresponds to ferrofluids with different concentrations of ferroparticles and the second one corresponds to solutions of Na_2SO_4 in water.

Amazing fact was found that there are two groups of points for ferrofluid and binary molecular mixture.

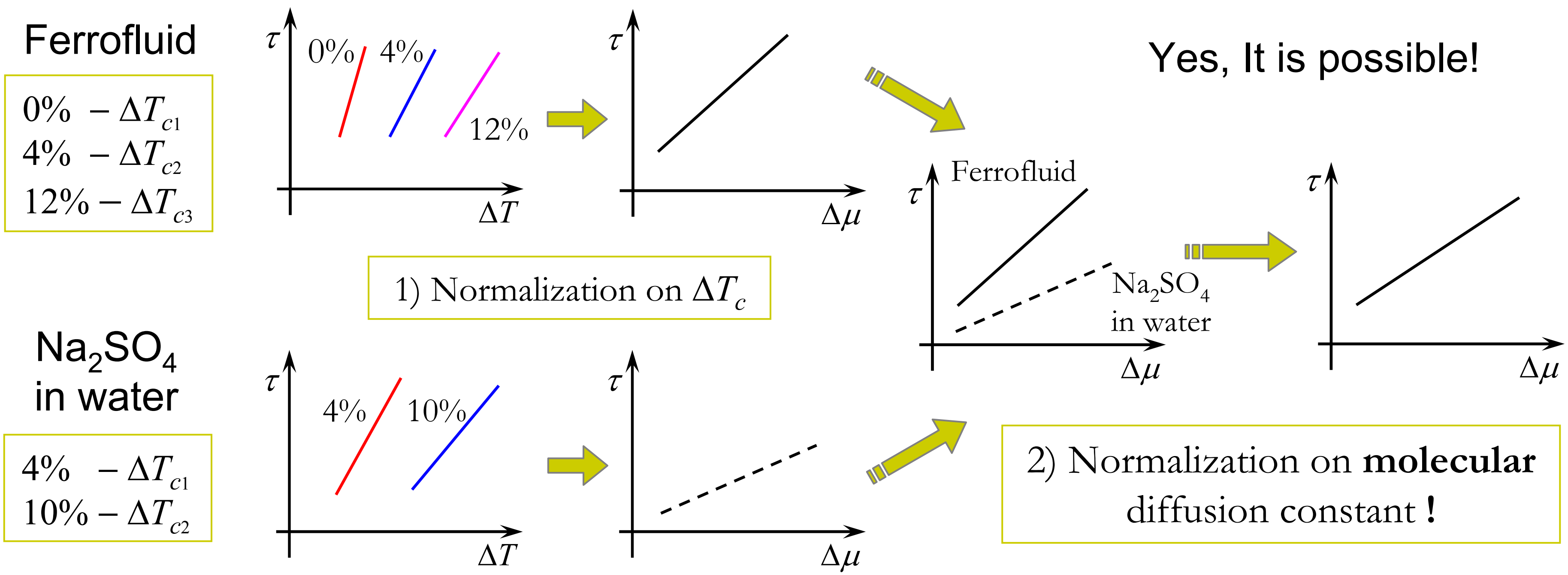
There is no visible dependence of period on particles concentration for ferrofluids. The same behaviour is observed for solutions of Na_2SO_4 in water.

In limiting case with zero concentration of particles for “pure” kerosene the regime of periodical redirection of flow circulation exists and has the same period.

“Grand” Unification

At first there were three different lines for ferrofluids and two lines for solution of Na_2SO_4 in water. Normalization on ΔT_c permits to unify these groups of lines in two dependencies of period on supercriticality.

At once the question arises: “Is it possible to combine these two lines in a **“united law”**?



$\underline{D = 7.6\cdot10^{-6}\ cm^2/c}$ (diffusion constant for solution of Na_2SO_4 in water), $D_f = 0.19\cdot10^{-6}\ cm^2/c$ (diffusion constant for ferroparticles in kerosene), $\underline{D = 3.5\cdot10^{-6}\ cm^2/c}$ (effective diffusion constant for molecular components of kerosene).

Crucial experiments

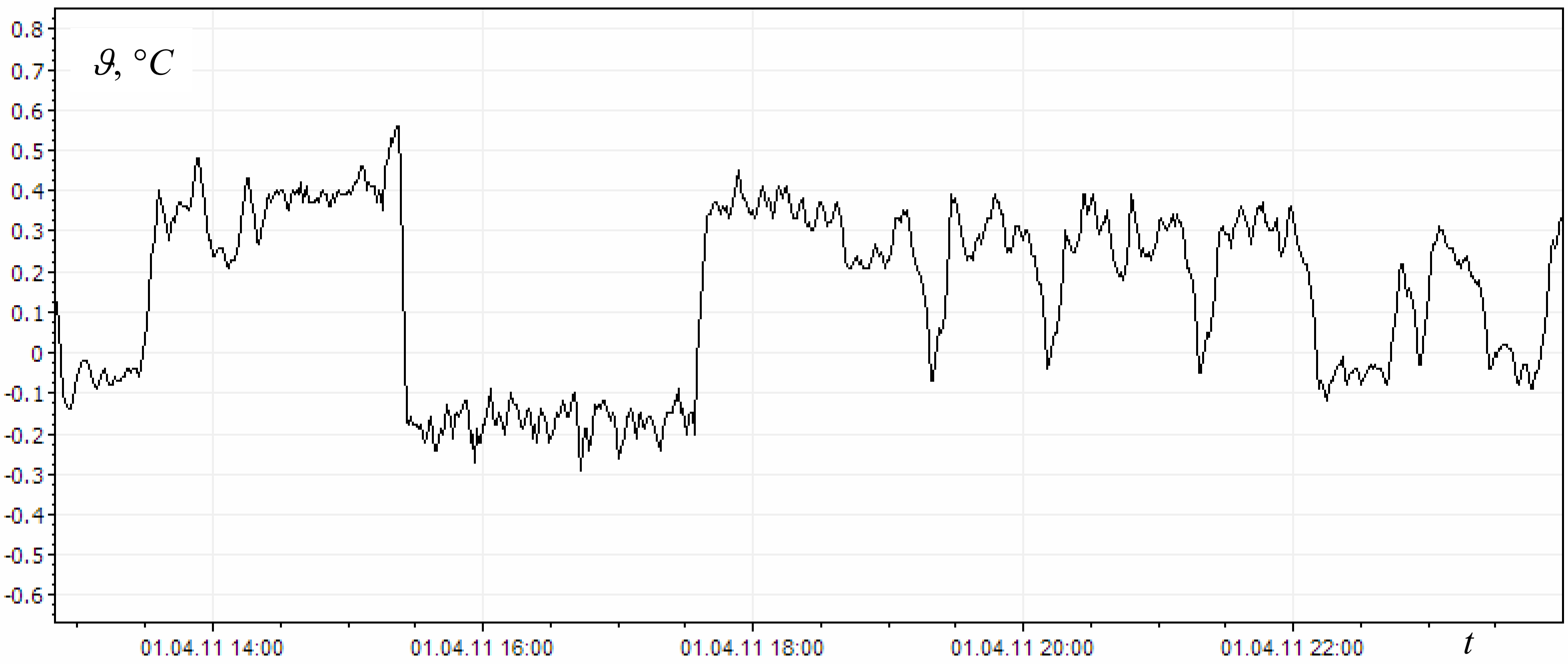


Fig. 11. Redirection of flow circulation is not regular because the critical temperature difference is very low. Flow characteristics become sensitive to small disturbances which cause spontaneous redirection of flow circulation .

- Spontaneous redirection of flow circulation takes place in “pure” **kerosene** and **diesel fuel**;
- Ferroparticles does not play key role in supporting of redirection of flow circulation;

Three component model of ferrofluid

State equation for density: $\rho = \rho_o(1 - \beta_t T' + \beta_c C' + \beta_\phi \phi')$

Equation of heat conduction:

$$\frac{\partial T}{\partial t} + (\vec{\mathrm{v}}\nabla)T = \frac{1}{\mathrm{Pr}}\Delta T$$

Equation for heavy molecular fraction in kerosene:

$$\frac{\partial C}{\partial t} + (\vec{\mathrm{v}}\nabla)C = \frac{1}{\mathrm{Sc}}\left(\Delta C + \varepsilon \Delta T\right)$$

Navies – Stokes equation:

$$\frac{\partial \vec{\mathrm{v}}}{\partial t} + (\vec{\mathrm{v}}\nabla)\vec{\mathrm{v}} = -\nabla p + \Delta \vec{\mathrm{v}} + \frac{\mathrm{Ra}H}{\mathrm{Pr}}(T - C - \phi)\vec{k}$$

Equation of particles transport:

$$\frac{\partial \phi}{\partial t} + (\vec{\mathrm{v}}\nabla)\phi = \frac{1}{\mathrm{Sc}_\phi}\left(\Delta\phi + \mathrm{Bl}\nabla\phi \cdot \vec{k}\right)$$

$$\mathrm{Bl} = \frac{\Delta\rho V_o g d}{k\bar{T}} = 0.02$$ - Boltzmann number

Basic boundary conditions:

Equilibrium state:

$$\begin{cases} T_o = -z/H \\ C_o = \varepsilon z/H \\ \phi_o = \tilde{\phi}_o e^{-\mathrm{Bl}z} \end{cases}$$

$$\left.\frac{\partial C}{\partial \vec{n}} + \varepsilon\frac{\partial T}{\partial \vec{n}}\right|_\Gamma = 0 \qquad \left.\frac{\partial \phi}{\partial \vec{n}}\right|_\Gamma = 0$$

Comparison of experiment with theoretical results

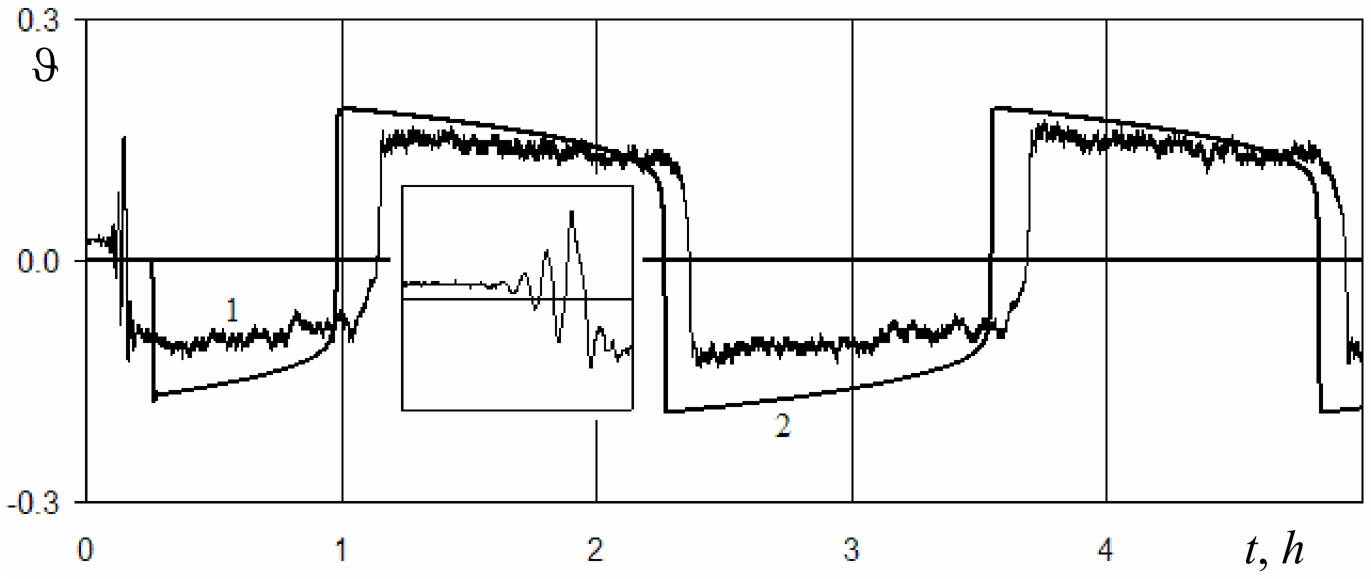


Fig. 12. Regime with redirection of flow circulation in ferrofluid;

a) *1* – experiment, MF **12%**; *2* – result of calculation for $\langle \phi \rangle = 0.3$, $\varepsilon = 0.01$, $H = 23$, Pr = 5.0, Sc = 16, $Sc_\phi = 60$, Bl = 0.02, $\xi_o = 10^{-5}$, $\Delta\mu = 1.14$.

b) Transitional oscillations (experiment) in dependence on time.

Comparison of experiment with theoretical results

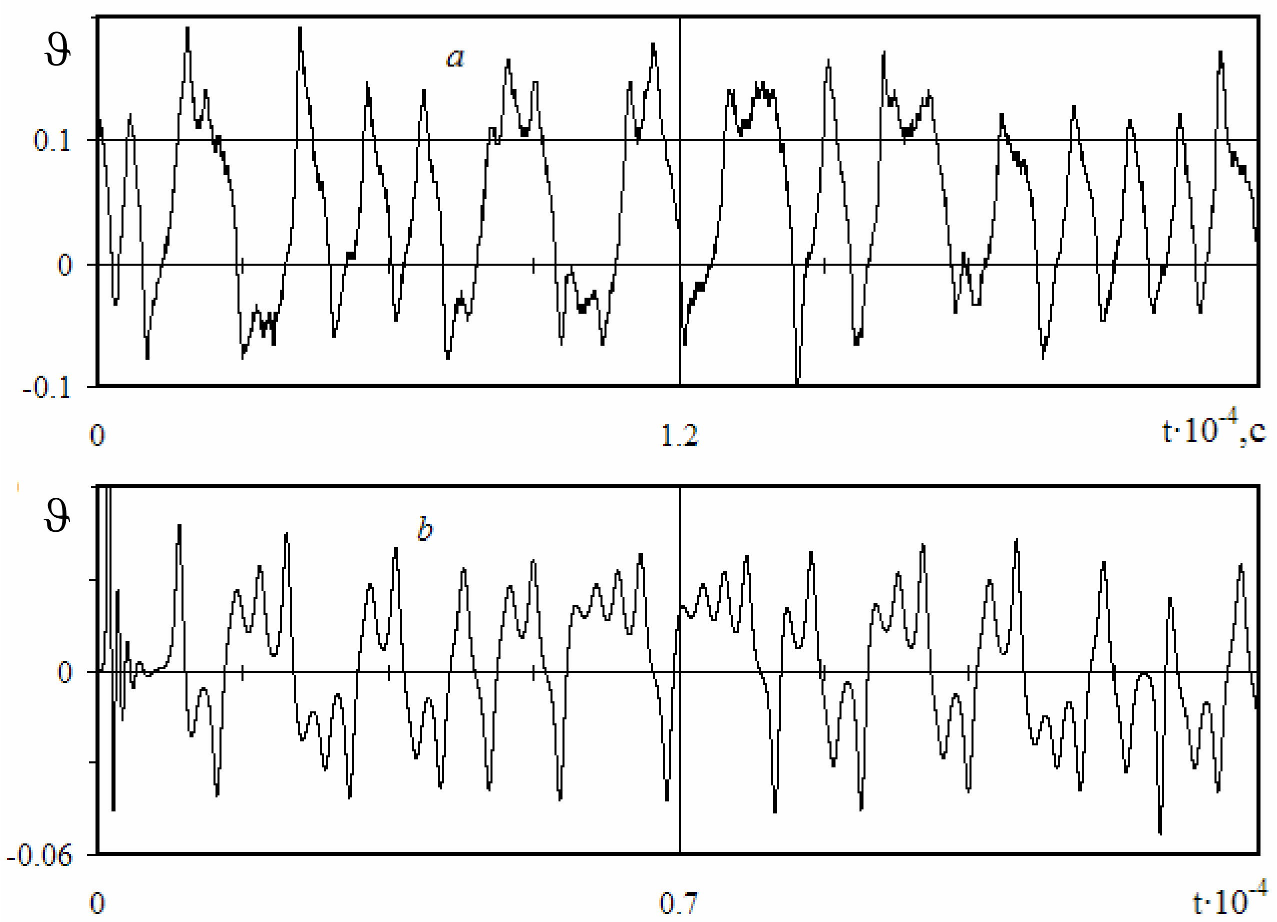


Fig. 13. Regime with irregular redirection of flow circulation in kerosene: *a* – experiment, *b* – theory

Normalized period of “flop-over” oscillations

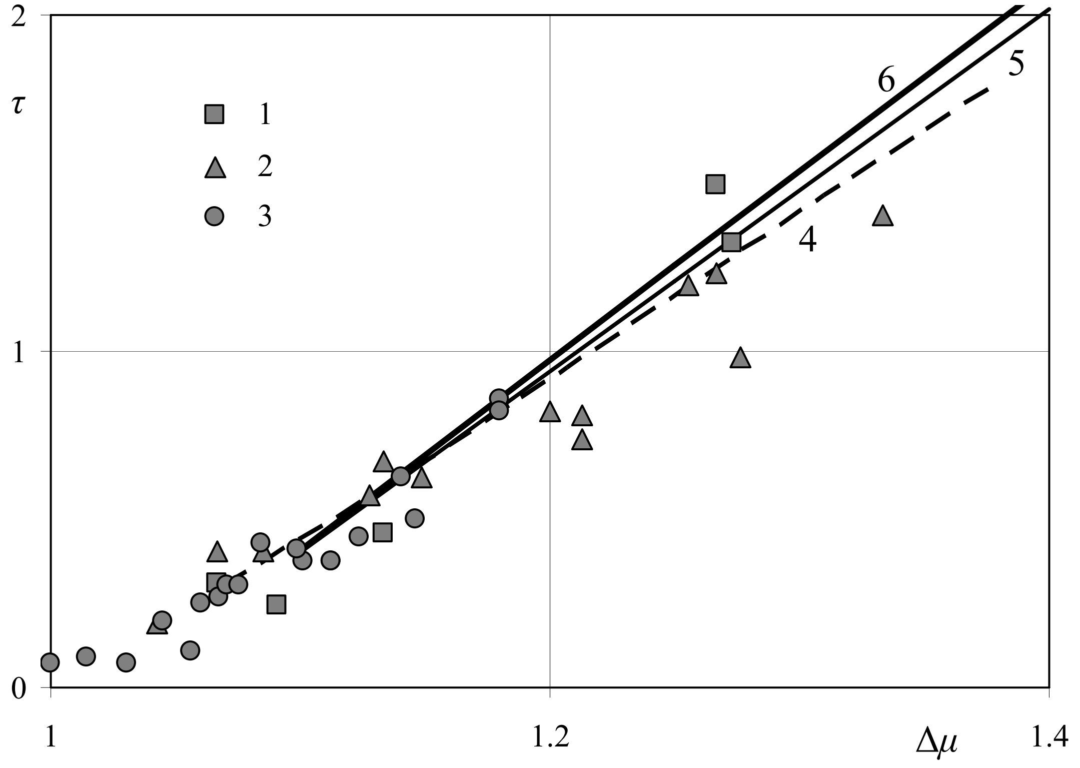


Fig. 14. Normalized period of regime with redirection of flow circulation in dependence on supercriticality: 1 - 3 – experiment; 1 – kerosene, 2 – ferrofluids with different concentrations of particles, 3 – solutions of Na_2SO_4 in water; 4 - 6 – calculation results: 4 – kerosene without particles, 5, 6 – ferrofluid with content of particles **4%**, **12%** (three component model).

Summary

❖ Thermal non-stationary convection of binary and multi-component liquid mixtures in connected channels with boundaries of high heat conductivity was investigated experimentally and theoretically. Experiments were carried out with the following mixtures: 1) carbon tetrachloride (CCl_4) in decane ($C_{10}H_{22}$), 2) aqueous solutions of sodium sulfate (Na_2SO_4), 3) water–ethanol mixtures, 4) magnetic fluid (stable colloidal suspension of ultra-fine ferromagnetic particles in kerosene).

❖ Over the threshold of convection specific "flop-over" oscillatory flows with very large period take place in the cases of binary molecular mixtures with normal thermodiffusion and magnetic fluids with different concentration of particles (4-12%). Direct numerical simulation on the base of hydrodynamics equations confirmed to results of experiments.

❖ Stationary convective flow settles in molecular mixtures with anomalous thermodiffusion that also was verified by the numerical calculations.

❖ Physical mechanisms were suggested to explain observed phenomena. According to our point of view the complex "flop-over" oscillatory regimes in binary molecular mixtures with normal thermodiffusion are determined by division of components in horizontal plane when the liquid moves predominantly along vertical heat-conducting boundaries of a cavity.

❖ Analogously to molecular mixtures periodic change of flow direction in magnetic fluid is explained by molecular thermodiffusion of kerosene components and depends on week effect of particles sedimentation.

Principal results

1) Molecular thermodiffusion is the main mechanism of restoring force origin that causes redirection of ferrofluid circulation in connected channels;

2) Three component model of ferrofluid was suggested to explain convective phenomena in thing channels;

3) During numerical modeling there was no reason to take into account the effect of particles thermodiffusion or other debatable effects to describe the experiments with magnetic colloids.

Acknowledgements:

- ✓ to the former governor of Perm region Oleg Chirkunov for the regular financial support of professors in our institutes of high education;
- ✓ to my wife which is always near as my rib.