

On the unsteady Reynolds thermal transpiration law

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Abstract. This paper presents the phenomenon of unsteady Reynolds thermal transpiration flow. The possible constitutive equations in the transpiration shell-like layer were studied analytically and numerically. There has been also examined experimental case of helium flow from cold to hot reservoir in nanopipe.

1. Introduction

Since 1879, when Reynolds has discovered the phenomena of thermal transpiration, this issue is still theoretically analyzed. Thermal transpiration can be defined as the macroscopic movement induced in a rarefied gas by a wall temperature gradient. The thermal mobility force, responsible for thermal transpiration, can be classified as a typical Newtonian “*vis impressa*” – a force which can be able to accelerate any massive substances along the wall [2]. In the literature there are still developed phenomenological models of the slip-regime for high Knudsen numbers [3,4], which can be also fitted to thermal transpiration for the case of gas in highly rarefied conditions, namely, in the free molecular regime. Hence, there is a need to prepare numerical implementation of Reynolds model of thermal transpiration at the contemporary experiment for example conducted by Rojas-Cárdenas et al [5].

In this paper, basing on experience in modelling of various surface phenomena [2-4,6-10] and starting from the first concepts by Reynolds [1], we have developed a consistent theory of thermal transpiration, that is able to describe thermal mobility both in rare and dense gases moving on an arbitrary solid surfaces. In particular, we discussed possible constitutive equations in the transpiration shell-like layer. The proposed model has been implemented and numerically assured by simulation of unstationary thermal transpiration benchmark experiment made by Rojas-Cárdenas et al [5].

1.1. Motivation

The *thermal transpiration* is not any pure academic problem and it has been recently used to construct the micro-electro-mechanical systems (MEMS) and nano-electro-mechanical systems (NEMS). The increased pumping effect could be obtained by a steeper gradient of temperature, extending tube length or making stages of pumping. Other micro-devices, which are based on both thermal transpiration and adherence slipping, called accommodation pumping, have been developed in nanotechnology [4].

Additionally, both recent developments in miniaturization and nanotechnology, as well as fullness unrecognized phenomena's occurring on a fluid-solid inter-phase within micro- and nano-scale compel us to reformulate conventional solid and fluid modelling. In nearby dividing surface, new inter-atomic forces make transport phenomena behave differently from their macroscopic counterparts. The most important ones are surface tension, surface curvature and surface twist that



possess quite different elastic properties [2-4]. Yet other one is generalized slip described by the Duhem, Navier, and du Buat numbers [7-10].

Owing to fast progress of nano-fluidic technology the nature of boundary conditions of fluid at solid surfaces has been revisited for the recent ten years. The huge surface to volume ratio turned our attention for fundamental understanding of nature of “surface-type driving forces” and the variety of different “jump” phenomena like slipping, rolling, spin-slipping, surface mobility, surface friction, thermo-porosis, etc [2-4]. In a very tiny channel there is no place for typical bulk phenomena, since whole flow is dominated by surface processes [6-11]. Moreover, these surface processes, in general, lead to some enhancement of transport [mass, momentum, thermal energy, electric current] in comparison with the bulk classical transport. Thus, quite surprising phenomena, unobserved in the bulk, appears in a surface layer and if a cross-section of a channel or pore becomes relatively small. Mentioned enhancement appearance could be easily visible and measurable [6-7].

However in this paper, we focused on this surprising phenomenon that the thermal transpiration is in theoretical point of view. Our main assumption is that this phenomenon is completely governed by a contact interface between a hot surface and colder gas. This is a surface migration of gas induced in a pipe with a gradient of temperature along its surface. This surface migration goes from colder to hotter places of surface. Thus, from mathematical modeling viewpoint, the thermal transpiration should be understood as some kind of “boundary conditions”.

The notion of “boundary condition” traditionally comes from well understood vocabulary of continuous fluid mechanics but numerous examples of the literature shows, that it could be misleading sometimes. In such case we wanted to change a paradigmatic look on “boundary condition” by assuming that within a boundary layer there are some additional, physically distinguished and measurable quantities which are necessary to create new class of “constitutive equations”. It practically means that we need to define the constitutive equations not only in the fluid bulk but also on the fluid-solid surface. Since, in practice, “boundary conditions” are seen as auxiliary mathematical-type constraints that allow solving partial-differential equation in a bulk, in the paper, we shell resign from exposing of “boundary conditions” notion and instead of it we will introduce a notion of a “boundary layer” or, equivalently, “interface layer” with boundary forces.

From this viewpoint, the boundary conditions related to the bulk equation of fluid motion [linear momentum balance], becomes “the surface layer equation of motion”. In a solid-fluid surface it is necessary to make up the separate equation of motion. Besides of the classical bulk, wall stress also appears quit new quantities like: surface friction force, surface mobility force, surface flux of momentum, etc. [3,5].

A main hypothesis of our paper is that the thermal transpiration is an example of surface mobility force which undergoes an unclassical procedure of constitutive modeling. We postulate that the status of surface constitutive equations should be equal to the status of constitutive equations in the fluid bulk. As has been already mentioned the thermal mobility force can be classified as a typical Newtonian “*vis impressa*” – a force which can be able to accelerate any massive substances. The phenomenological models of thermal transpirations existing in the literature are based on different approaches to kinetic theory of gases [12], which are usually restricted to a very small calculation domain. The more consist one, proposed here, can be also fitted to slip-regime for high Knudsen numbers and, can be more correctly extended the so-called “second order models” [13,14].

2. Model description

In this paper, we have assumed that the continual motion of gas in a nano-channel is governed simultaneously by two kinds of equations of motion [8], mainly, the bulk equation of motion:

$$\partial_t(\rho \mathbf{v}) + \text{div}(\rho \mathbf{v} \otimes \mathbf{v} + \mathbf{p}) = \rho \mathbf{b} \quad (1)$$

and the surface layer equation of motion:

$$\partial_t(\rho_s \mathbf{v}_s) + \text{div}_s(\rho_s \mathbf{v}_s \otimes \mathbf{v}_{s\parallel}) + \text{div}_s(\mathbf{p}_s) - \varpi \mathbf{n} + \mathbf{p}\mathbf{n} + \mathbf{f}_{\partial V} = \rho_s \mathbf{b}_s \quad (2)$$

In above $\rho \mathbf{v}$ and $\rho_s \mathbf{v}_s$ are the bulk and the surface momentum density vectors, ρ and ρ_s are the gas density in the bulk and on the boundary, respectively. Next, having an original Reynolds' notations in mind, $\mathbf{p} = p_{ij} \mathbf{e}_i \otimes \mathbf{e}_j = \mathbf{p}^T$ and $\mathbf{p}_s = \mathbf{p}_s^T$ are the bulk and the surface flux of momentum, \mathbf{n} is the unit normal vector on the boundary surface, $\mathbf{f}_{\partial V}$ – is the boundary force, \mathbf{b} and \mathbf{b}_s are the body forces in the bulk and on the boundary. Additionally, ϖ is the Stokes normal surface pressure, due to assumption that in the boundary layer a usual spherical pressure tensor changes into an ellipsoidal pressure tensor. Surface divergence is defined as a right contraction of surface gradient: $\text{grad}_s(\cdot) = \text{grad}(\cdot) \mathbf{I}_s$. Tangent to surface component of the slip velocity is $\mathbf{v}_{s\parallel} = \mathbf{v}_s \mathbf{I}_s$ where the surface Gibbs identity is defined to be: $\mathbf{I}_s = \mathbf{I} - \mathbf{n} \otimes \mathbf{n}$ and generally \mathbf{v}_s - slip velocity is equal $\mathbf{v}_{sl} = \mathbf{v} - \mathbf{v}_{wall}$. Hence Poiseuille flow can be modeled by equation (1).

In the case, when we can omit whole dynamics of a surface layer, and the surface flux of momentum is represented only by the Young-Laplace surface tension: $\mathbf{p}_s = \gamma \mathbf{I}_s$, the surface equation of motion (2) reduces to the Stokes boundary condition [2]:

$$\text{div}_s(\gamma \mathbf{I}_s) - \varpi \mathbf{n} + \mathbf{f}_{\partial V} + \mathbf{p} \mathbf{n} = 0 \quad (3)$$

Let us note that this boundary condition acts independently from any constitutive definition of Stokes pressure tensor – it can be defined traditionally to be: $\mathbf{p} = p \mathbf{I} - 2\mu \mathbf{d} + 2/3 \mu I_d \mathbf{I}$ or it can also consist a transpiration part or a second order part. Hitherto, we denote the rate of deformation as: $\mathbf{d} = 1/2(\text{grad } \mathbf{v} + \text{grad}^T \mathbf{v})$; $I_d = \text{tr}(\mathbf{d})$.

2.1. The boundary forces

The boundary force, responsible for a so-called generalized slip can be splinted on two contradictive components: surface friction and surface mobility: $\mathbf{f}_{\partial V} = \mathbf{f}_r + \mathbf{f}_m$. Both forces are the subject of constitutive modeling. For instance, the friction force possesses three known contributions: Duhem, Navier and du Buat [4]:

$$\mathbf{f}_r = \nu_0(p - \varpi) \mathbf{e}_f + \nu_1(\mathbf{v} - \mathbf{v}_{wall}) + \nu_2(\mathbf{v} - \mathbf{v}_{wall}) \mathbf{e}_f \quad (4)$$

where slip versor is defined as: $\mathbf{e}_f = (\mathbf{v} - \mathbf{v}_{wall}) / |\mathbf{v} - \mathbf{v}_{wall}|$ and ν_0 ; ν_1 ; ν_2 are three friction coefficients [Duhem, Navier, du Buat, respectively] which are simultaneously under influence of the material of fluid and solid surface. However the surface mobility forces, which act usually against a surface friction, describe surface contributions from a numerous transpiration phenomena [2]:

$$\mathbf{f}_m = -c_{mp} \text{grad}_s(p - \varpi) - c_{m\theta} \text{grad}_s(\theta) - c_{mN} \text{grad}_s(N) - \dots \quad (5)$$

which depends linearly on surface gradient of surface pressure, surface temperature and surface concentration. More information is in section about mobility force reconstruction. In particular, the main goal of the paper is to discuss only Reynolds' contributions to definition of surface mobility modeling: $-c_{m\theta} \text{grad}_s \theta$.

2.2. Thermal transpiration discovery

Osborne Reynolds is the one author, who entirely devoted to the discovery of thermal transpiration [1]. He asserts that a primary reason of thermal transpiration in the bulk motion is not the second gradient of temperature along axes of a capillary, but the axial gradient of acceleration that acts close to the wall surface it makes the enhancement of the normal Navier slip.

Let us now return, to the paper [1], mainly concerning with the experimental discovery of the thermal transpiration and its description by a Maxwellian-like kinetic theory of transpired gases.

Reynolds unified three phenomena: the Graham *pressure transpiration*, the Graham *concentration transpiration* [15] and, just discovered by Reynolds, the *thermal transpiration* became into a one generalized model of transpiration phenomena.

Thermal transpiration was originally discovered in thin (5.3-6.3 mm) porous plates made of meerschaum, stucco or graphite. Reynolds has observed that there is the difference in rates at which different gases transpire through minuscule pore channels for the same initial pressure and the same difference of temperature on two sides of plate. Simultaneously, the changing of initial pressure on both sides of the plate was observed. After some time (equilibrium time) the difference of pressure between the hot and cold sides becomes constant and no more migration of gas is observed. It means that the difference of temperature on the two sides of porous plate might cause gas, without any initial difference of pressure to pass through the plate from the cold to the hot side.

For instance, the permanent (equilibrium) difference of pressure equal to an inch of mercury and the higher pressure on the hotter side is achieved in the atmospheric pressure, a difference 160 °F (from 52 to 212 °F) with hydrogen on both sides of the stucco plate of thickness 6.3 mm. Moreover, Reynolds has found that a wall gradient of temperature leads to different mass flow of different gases. This effect can be potentially used as a separation mechanism for gas mixtures [5].

Summarizing the results of his experiments, Reynolds has deduced eight first phenomenological laws of the thermal transpiration. Nowadays, the most known of them is, the “Law IV”, which says that in the equilibrium, for those cases when rarefaction has reached the point at which the difference of pressure is nearly proportional to the density, the ratio of hot and cold gas pressures is in square roots to the absolute temperature:

$$\frac{p_H - p_C}{p_H} = \frac{\sqrt{\tau_H} - \sqrt{\tau_C}}{\sqrt{\tau_H}} \quad (6)$$

Let us note, that in both – meerschaum and stucco plates of 6.3 mm thick, it is tendency of the pressure difference $\delta p = p_H - p_C$, for hydrogen, air and carbon acid [see[1]; original Figures 4,5,6], as the initial pressure rises, to a maximum value. The indices *H* (hot), *C* (cold) have appeared in Reynolds’ paper only just in further part of theoretical considerations. Note that denotation of the absolute temperature (τ) coming from Rankine’s proposal.

2.3. Thermal mobility force reconstruction

In order to show how Reynolds made his model of thermal transpiration one should follows in details of Reynolds construction of kinetic theory of rarefied gases – in the bulk and on a surface. Original Reynolds’ line of reasoning is based on his own previous achievements concerning forces which must result when “heat” is communicated from a solid surface to a gas. Reynolds’ model of special could define of tangential and normal forces, especially when small spaces might be allowed for the gas molecules to pass. Moreover, Reynolds was able to show that it is a “complete theory of transpiration phenomena” which describes not only thermal transpiration but any other transpiration phenomena discovered by Graham [15], for instance a phenomena called “impulsion”.

Reynolds’ denotations - ϖ, θ, N - means the normal pressure, temperature and mole concentration of a gaseous component. Hence, we could a unified model of pressure, temperature and concentration transpirations on the boundary layer, by adding particular contributions described by equations: [see [1]: original eqs.:(110)(112)(117)], respectively. Now, reducing eq. (2) only to so-called Stokes’ boundary layer equation $\mathbf{f}_{\partial V} - \varpi \mathbf{n} + p \mathbf{n} = 0$, the Reynolds model of slip-mobility layer can be reconstructed shortly to be:

$$-c_{m\varpi} \text{grad}_s \varpi - c_{m\theta} \text{grad}_s \theta - c_{mN} \text{grad}_s N + \nu(\mathbf{v} - \mathbf{v}_{wall}) + (p - \varpi)\mathbf{n} - 2\mu \mathbf{d}\mathbf{n} + 2\beta \mathbf{d}_{(2)}\mathbf{n} = 0 \quad (7)$$

Realizing the Graham phenomena, the appearing of pressure transpiration gradient is explicit in the model. The pressure mobility coefficient $c_{m\varpi}$ is different for any gases in the same material of capillary tube. The definition of the thermal mobility coefficient $c_{m\theta}$ where Reynolds has introduced four accommodation coefficients f : mainly f_1, f_2, f_3, f_4 - two first depend, separately, on the properties of solid surface material and a kind of gas comes across in this model. Next, we are going to describe two processes of interaction of a gas with a boundary. The diffusion transpiration of Graham is defined by the concentration mobility coefficient c_{mN} . About β and $\mathbf{d}_{(2)}$ next section presents.

2.4. Consistency of thermal transpiration with a 3D model of momentum transport

It should be added that the definition of the Cauchy momentum flux on the boundary - \mathbf{p} contains Reynolds' proposition extends a definition of \mathbf{p} taking the diade of acceleration of deformation $\mathbf{d}_{(2)}$. Reynolds, further applied a coefficient β responsible for accelerative contributions, it should be find on the base of a previous researches of "dimensional properties of mater".

The consistency of mathematical model of fluid in a bulk and within the layer boundary was the reason for writing the paper [16]. Here, Reynolds proposes a new definition of the Cauchy momentum flux, where it appears an *acceleration of deformation diade* - $\mathbf{d}_{(2)}$, defined as a symmetric tensor:

$$\mathbf{d}_{(2)} = 1/2 (\text{grad } \mathbf{a} + \text{grad}^T \mathbf{a}) \neq \dot{\mathbf{d}} . \quad (8)$$

Thus, the Stokes form of the momentum flux is extended to be:

$$\mathbf{p} = p\mathbf{I} - 2\mu \mathbf{d} + \frac{2}{3} \mu \mathbf{I}_d \mathbf{I} + 2\beta \mathbf{d}_{(2)} \quad (9)$$

where, the Reynolds coefficient $\beta = r/p$ is inversely proportional to pressure. Hence, substituting the above constitutive equation to the momentum balance $\rho \dot{\mathbf{v}} + \text{div } \mathbf{p} = \rho \mathbf{b}$, and omitting $\text{div}(\text{grad}^T \mathbf{a}) \approx 0$, we obtain after Reynolds [1, eq.(12)]:

$$\rho \mathbf{a} + \beta \text{lap} \mathbf{a} + \text{grad } p - \mu \text{lap } \mathbf{v} - \frac{1}{3} \mu \text{grad } \mathbf{I}_d = \rho \mathbf{b} . \quad (10)$$

The part with β coefficient can be further omitted in the bulk domain of fluid flow, but on the boundary it represents very important - an accelerative - contribution to the slip conditions.

2.5. Law of mass flow rate in a circular tube

Finally, Reynolds has combined Poiseuille pressure driven flow with transpiration pressure driven flow showed in eq (10) by adding the transpiration mobility force (5). To find a formula describing an average velocity in a long pore on a diameter a , it was necessary to assume that characteristic mean-free path of the gas molecules, say l , is the same range as the geometrical parameter a . Then the mass flow rate is [1, eq. 101]:

$$\dot{m} = \rho(\pi a^2) v_z^{aver} = -\rho(\pi a^2) a \sqrt{\frac{2p}{\rho}} \left\{ \mathcal{F}_1(a/l) \frac{1}{p} \frac{dp}{dz} - \mathcal{F}_2(a/l) \frac{1}{\theta} \frac{d\theta}{dz} \right\} . \quad (11)$$

If $\dot{m} = 0$ then the net motion vanish and some constant proportion between $dp/d\theta$ is obtained. The time needed to reach $\delta p = p_H - p_C = \text{const}$ is easy to be measured. Appearing above function $\mathcal{F}_1, \mathcal{F}_2$ are not only the function of (a/l) but also of four accommodation coefficients f_1, f_2, f_3, f_4 .

2.6. Simplified Reynolds' model of a boundary layer with the thermal mobility

Despite the fact that investigation on the thermal transpiration has been started long time ago [see: 13,17] many questions remain up to now unsolved. This is connected with a special feature of the thermal transpiration, which is very sensitive to a kind of solid material and a kind of rarified gas. In this paper, thermal transpiration mobility force has been modeled as:

$$\mathbf{f}_m = -c_{m\theta} \text{grad}_s \theta. \quad (12)$$

Contrary, mass flux proposed by Reynolds is, in general, sensitive, both, on the properties of gas and solid surface and depends on two *thermal momentum accommodation coefficients* f_3, f_4 . However, having a complete experimental data, for instance works of Rojas-Cárdenas et al. [5,18,19], Daggerbart [17], Reese et al. [12], Ewart et al. [20], Pitakarnnop et al. [21], one tray to develop a consistent closure to the $c_{m\theta}$ coefficient within the framework of continuum modeling.

3. Analysis

It should be mentioned that the above cited law (eq 6) had validity only at a zero-flow $\dot{m} = 0$ final equilibrium state, far from transitional stage of gas displacement, especially at a point where $\dot{m} = \max$. Additionally Daggerhart [17] proposed semi-empirical formula that goes over $p_H / p_C = (\theta_H / \theta_C)^{1/2}$ and predict how the thermo-molecular pressure ratio p_H / p_C at the final equilibrium zero-flow state varies in function of the gas rarefaction, the gas physical properties, and the applied temperature difference to the tube. Many various papers are discussed and verified by Reese, Zheng, and Lockerby [12] as well as by Storvick, Park and Loyalka [13] however a new benchmark experiment has appears in 2011 made by Rojas-Cárdenas et al. [5] and was extended in another works [18,19].

3.1. Geometry and boundary conditions

Rojas-Cárdenas et al [5] have applied an original method for thermal transpiration induced mass flow rate measurements conducted via measuring *in situ* the pressure evolution in time at both ends of the tube using two high-speed response pressure gauges. In the long circular cross-section glass (borosilicate) microtube $D=2R=490 \mu\text{m}$; $L=3.053 \text{ cm}$ is connected to two reservoirs (Fig.1): cold (no 1, environmental temperature) and hot (no 2, $\theta_2 = 80^\circ \text{C}$, heated by an internal heater) (Fig.1) with volume ratio of the two reservoirs $V_H / V_C = 0.81$. Before experiment starts, the pressure inside the both reservoirs was regulated by means the vacuum system and stay between 13.3 and 1330 Pa for helium. For numerical simulations we have chose $V_1 = V_C = 14.85 \text{ cm}^3$.

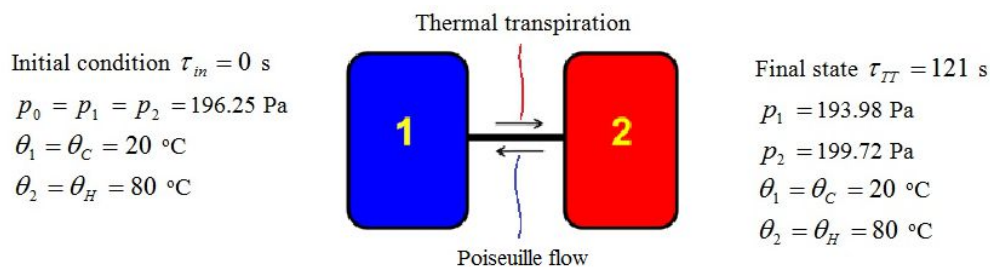


Figure 1. Scheme of helium flow from cold (1) to hot (2) reservoir Rojas-Cárdenas et al [5].

After implementation of mobility force (12) into a commercial code, the Rojas-Cárdenas et al benchmark experiment [5] has been preformed for a basic data corresponding to helium. For numerical simulation we have taken the following data: initial pressure $p_0 = p_{in} = 196.25 \text{ [Pa]}$, cold

temperature $\theta_C = 20$ [°C], hot temperature $\theta_H = 80$ [°C] (with linear distribution along the micro-pipe), referential viscosity $\mu = 1.96 \times 10^{-5}$ [Pas], individual gas constant $\tilde{R} = 2077$ [J/kgK], the ratio of specific heats $\kappa = 1.667$.

Basing on our previous considerations [3,4,7,8] and having in mind proposed in the literature value of thermal accommodation coefficient [12,18,19], we have made a calibration of $c_{m\theta}$ value. Standing on Rojas-Cárdenas et al data [5] and Ewart et al considerations [20], this coefficient is defined as follows:

$$c_{m\theta} = \frac{3}{4} \frac{\mu \nu}{\rho \theta} = \frac{3}{4} \frac{\mu^2}{\rho \theta l_s} \quad (13)$$

The main unknown was the Navier slip viscosity ν that can be defined by slip length also [2-4]. We assume that numerical value of $c_{m\theta}$ has been calculated using the definition the Helmholtz-Piotrowski slip length: $l_s = \mu/\nu$, and helium-glass slip length $l_s = 0.00016$ cm. In eq (13) the temperature along the micro-pipe was taken according to linear distribution.

4. Results and discussion

Opening a disconnecting valve the flow induced by thermal transpiration makes pressure in both reservoirs equal and flow from cold to hot places derived only by the wall mobility force (2), that depends on $c_{m\theta}$ value between glass and helium and the wall temperature gradient. The pressure in the cold decrease and at the hot palace increases (Fig. 2a). Now, thermal transpiration forces achieve maximal value and mass flux attain pick in a characteristic time τ_R (Fig. 2b).

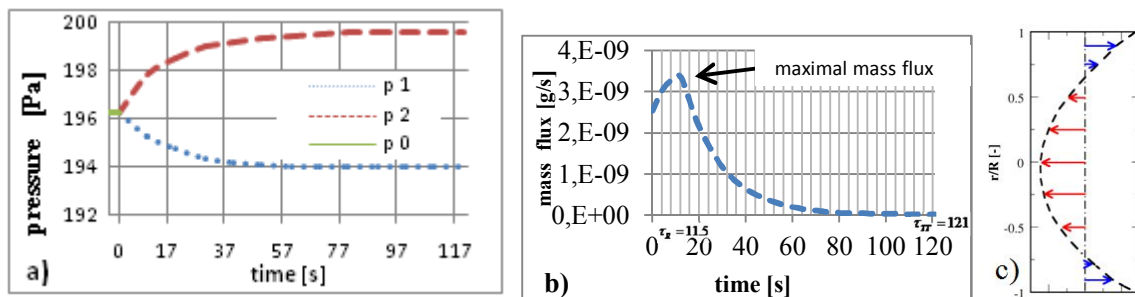


Figure 2. Results of unsteady thermal transpiration modeling of the Rojas-Cárdenas et al benchmark experiment [5] with the thermal transpiration mobility force (eq. 2): a) plot of pressure change in (2) and (1) reservoir; b) the plot of mass flux c) stationary velocity profile.

After that we included the Poiseuille flow governed by difference of pressure (Fig. 2a), that finally leads, after time τ_{TT} to equilibrate state when the pressure difference arrives to its maximum and the resulting mass flux is zero. Our results agreed with the experiment [5]. It is easy to numerically prove that at the final equilibrium zero-flow state (after 121 seconds) is connection of Poiseuille flow in the centre of channel and counter thermal transpiration flow on the surface (Fig. 2c).

5. Conclusions

Reassuming, the numerical implementation of Reynolds model of thermal transpiration and its usefulness for description of an experiment by Rojas-Cárdenas et al [5] have been performed. Finally, Poiseuille flow in the centre of channel and counter thermal transpiration flow on the surface has been achieved.

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