

PARAMETRIC STUDY OF FLUID DYNAMICS IN PEM FUEL CELLS

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This paper is a research in the frame of the Grant CEx 189/2006, “Water and thermic management for PEM fuel cells”. We study here the flow in the “Gas Diffusion Layer” of a PEM fuel cell. In this region, the dynamic of fluids is governed by capillary and diffusion phenomenon. The diffusion-convection process is studied, involving the oxygen and water vapors. Our model is similar with the Hele-Shaw model, which describes the displacement of two immiscible fluids with different viscosities between two parallel plates at a small distance (see [19]). The concentrations of the fluids are invertible in terms of the viscosities. If the displacing fluid is less viscous, the interface between the fluids is unstable. We consider a diffusion region between the two fluids, where the viscosity is a parameter. We study the linear stability of a basic solution, in terms of the diffusion coefficient. We get an upper estimate of the growth constant of perturbation. A large enough diffusion coefficient gives us a significant improvement of the flow stability.

Key words: Proton exchange membrane, Fuell cells, Numerical methods, Eigenvalue problems

1. INTRODUCTION

The water transport in a PEM fuel cells (PEMFC) is governed by the mass transfer and diffusion appearing in the “Gas Diffusion Layer” (GDL) region. Some previous and interesting results are given in [3], [4], [5], [13]. A capillary pressure exists on the interface gas-liquid and the surface tension is involved. In [3], the capillary pressure is given by Genuchten’s function, described also in [14]. The diffusion process is characterized by the diffusion coefficient η . In [4] are given some methods to find η .

At the Workshop “Modeling and Simulation of PEM Fuel Cells”, Freiburg Universität, 2006, are studied some diffusion and transport phenomena in PEMFC. In [15] is studied the transport in the catalize layer; in [17] is studied the bi-phasic flow in complex porous media; in [1] are estimated the effective diffusivity coefficients. The homogenization method is used in [18] to obtain a reduced model of GDL. A triphasic model is studied in [2]. All these papers are related to our approach.

Some qualitative and numerical aspects, related to the transfer processes PEM, are studied in [6]-[7] and [8]-[12].

In this paper, we consider a quasi-one dimensional model for the GDL. The length is much longer compared with the thickness. In [13], at the ends are used periodicity conditions; then in fact our medium can be considered of infinite length in the displacement direction. On this way, we can use the Hele-Shaw model. If displacing fluid (gas) is less viscous, the interface with the displaced fluid (water) is unstable. Between the two fluids we consider a “Transition Zone” (TZ), where the viscosity is an increasing function. We have two interfaces, with two surfaces tensions; here we have small jumps of the viscosities. In TZ we allow a diffusion process. A basic steady solution, with straight initial interfaces, is considered. We study the linear stability of this basic solution. We obtain a qualitative result concerning the improvement of the stability and an upper estimate of the growth constant. A large enough diffusion coefficient η gives us an improved stability.

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2 THE HELE-SHAW MODEL

In [16] is considered the flow of a Stokes fluid between two parallel plates at a small distance δ , in the plane xOy ; Oz is orthogonal on the plane. In the following, (u, v, w) , p are the velocity and the pressure. In our case $w = 0$, then p is not depending on z . The x, y derivatives of the velocity are neglected, compared with the z derivative. Therefore we get the system

$$\mu \frac{\partial^2 u}{\partial z^2} = \frac{\partial p}{\partial x}, \quad \mu \frac{\partial^2 v}{\partial z^2} = \frac{\partial p}{\partial y},$$

where μ is the viscosity. A no-slip condition is imposed at $z=0$, $z=\delta$, then

$$\bar{u} = -\frac{\delta^2}{12} \frac{1}{\mu} \frac{\partial p}{\partial x}, \quad \bar{v} = -\frac{\delta^2}{12} \frac{1}{\mu} \frac{\partial p}{\partial y} \quad (\text{A1})$$

$$\bar{u} = \frac{1}{\delta} \int_0^\delta u(x, y, z) dz, \quad \bar{v} = \frac{1}{\delta} \int_0^\delta v(x, y, z) dz.$$

The above equation is obtained by using Taylor expansion of second order for u, v . The relation (A1) is quite similar with the Darcy law, which governs the flow in a porous two-dimensional medium, with the permeability $\delta^2/12$. The filtration velocity is given by \bar{u} and \bar{v} . We emphasize that only the permeability and velocity are ‘‘fictive’’, while the viscosity in (A1) is the same as in the Darcy law.

We conclude that, in thin plane regions (in our case the domain between two plates), the flow of a viscous fluid is governed, with a good approximation, by the Darcy law in a fictive porous medium.

The fuel cells are ‘‘composed’’ by some thin plane regions, where the flow and transfer processes are performed. The above considerations allow us to approximate the flow in these regions by the Darcy law, in the frame of the Hele-Shaw model.

3. THE MATHEMATIC MODEL OF DIFFUSION IN GDL

In the fixed plane x_1Oy , we consider the flow of three fluids in a homogeneous porous medium, using the Hele-Shaw model. The medium is saturated with following three fluids: oxygen-vapors, a mixture formed by oxygen-vapors and water, and the third part filled with water. We have three viscosities: oxygen vapors with constant viscosity μ_1 , the mixture with variable viscosity μ , and water with constant viscosity μ_2 . All regions are moving by oxygen velocity U far upstream. We have also two interfaces, denoted by $\Gamma_i(x_1)$.

The flow is governed by the Darcy law, the continuity equation for velocities and a ‘‘conservation’’ law for the viscosities in the mixture (here the viscosities replaced the concentrations, as we mentioned above).

Let (u, v) and p be the velocity and the pressure. The temperature is considered constant and we study only the hydrodynamical aspects. The case of a variable temperature was considered in a previous study, related to the same Grant.

We neglect the adsorption and dispersion phenomena. Therefore the flow is governed by the following equations:

$$\frac{\partial u}{\partial x_1} + \frac{\partial v}{\partial y} = 0, \quad \frac{\partial p}{\partial x_1} = -\mu u, \quad \frac{\partial p}{\partial y} = -\mu v, \quad x_1 \notin \Gamma_1(x_1) \cup \Gamma_2(x_1); \quad (1)$$

$$\frac{\partial \mu}{\partial t} + u \frac{\partial \mu}{\partial x_1} + v \frac{\partial \mu}{\partial y} = \eta \left\{ \frac{\partial^2 \mu}{\partial x_1^2} + \frac{\partial^2 \mu}{\partial y^2} \right\}, \quad x_1 \in (\Gamma_1(x_1), \Gamma_2(x_1)) \quad (2)$$

$$\mu(\Gamma_1(x_1)) = \mu_1, \quad \mu(\Gamma_2(x_1)) = \mu_0 < \mu_2; \quad (3)$$

$$\mu = \mu_1, \quad x_1 < \Gamma_1(x_1); \quad \mu = \mu_2, \quad x_1 > \Gamma_2(x_1). \quad (4)$$

On $\Gamma_i(x_1)$ we use the Laplace law: the pressure jump is balanced by the surface tension multiplied with the interface's curvature. The normal component of the velocity on the interfaces $\Gamma_i(x_1)$ is continuous. Far from $\Gamma_i(x_1)$ we have the condition

$$u = U, v = 0, x_1 \ll \Gamma_1(x_1) \text{ or } x_1 \gg \Gamma_2(x_1). \quad (5)$$

The following basic solution (6)--(10) exists for the problem (1)--(5):

$$u = U, v = 0, \frac{\partial P}{\partial x_1} = -\mu U, \frac{\partial P}{\partial y} = -\mu v, x_1 \notin \Gamma_1(x_1) \cup \Gamma_2(x_1), \quad (6)$$

$$\Gamma_1(x_1) : x_1 = Ut - l, \Gamma_2(x_1) : x_1 = Ut, \quad (7)$$

$$\frac{\partial \mu}{\partial t} + U \frac{\partial \mu}{\partial x_1} = \eta \frac{\partial^2 \mu}{\partial x_1^2}, x_1 \in (\Gamma_1(x_1), \Gamma_2(x_1)); \quad (8)$$

$$\mu = a(x_1 - Ut) + b, x_1 \in (\Gamma_1(x_1), \Gamma_2(x_1)) \quad (9i)$$

$$\mu |_{\Gamma_1(x_1)} = \mu_1, \mu |_{\Gamma_2(x_1)} = \mu_0. \quad (9ii)$$

The viscosity in the mixture is a linear function in terms of $(x_1 - Ut)$. We have two straight material interfaces, then the following condition holds:

$$P \text{ is continuous on } \Gamma_i(x_1). \quad (10)$$

Our task is to study the linear stability of the *basic solution* (6)--(10).

4. THE STABILITY SYSTEM

We consider the perturbations u', v', p', μ' of the basic velocity, pressure and viscosity and get the system

$$\begin{aligned} \frac{\partial(U + u')}{\partial x_1} + \frac{\partial v'}{\partial y} &= 0, x_1 \notin \Gamma_1(x_1) \cup \Gamma_2(x_1); \\ \frac{\partial(P + p')}{\partial x_1} &= -(\mu + \mu')(U + u'), \frac{\partial(P + p')}{\partial y} = -(\mu + \mu')v', x_1 \notin \Gamma_1(x_1) \cup \Gamma_2(x_1); \\ \frac{\partial(\mu + \mu')}{\partial t} + (U + u') \frac{\partial(\mu + \mu')}{\partial x_1} &= \eta \frac{\partial^2(\mu + \mu')}{\partial x_1^2}, x_1 \in (\Gamma_1(x_1), \Gamma_2(x_1)), \end{aligned}$$

with the following boundary conditions

$$\text{a) the Laplace's law on } \Gamma_i(x_1); \text{ b) } u', v' = 0 \text{ far from } \Gamma_i(x_1); \text{ c) } \underline{\mu' = 0 \text{ on } \Gamma_i(x_1)}. \quad (11)$$

We use the *mobile* coordinate system (x, τ) :

$$x = x_1 - Ut, \tau = t$$

therefore for the arbitrary function $F(x, y)$ we get the relation

$$\frac{\partial F}{\partial t} + U \frac{\partial F}{\partial x_1} = \frac{\partial F}{\partial \tau}$$

However, in the following, τ is also denoted by t . In the *mobile* system, the perturbations are governed by the problem

$$\frac{\partial u'}{\partial x} + \frac{\partial v'}{\partial y} = 0, x \notin \{-l, 0\}, \quad (12)$$

$$\frac{\partial p'}{\partial x} = -\mu u' - \mu' U, x \notin \{-l, 0\}, \quad (13)$$

$$\frac{\partial p'}{\partial y} = -\mu v', x \notin \{-l, 0\}, \quad (14)$$

$$\partial \mu' / \partial t + u' \partial \mu' / \partial x = \eta \left\{ \partial^2 \mu' / \partial x^2 + \partial^2 \mu' / \partial y^2 \right\}, x \notin (-l, 0), \quad (15i)$$

$$\mu = ax + b, \quad \mu(-l) = \mu_1, \quad \mu(0) = \mu_0 < \mu_2, \quad x \in (-l, 0), \quad (15ii)$$

$$\mu = \mu_1, \quad x < -l, \quad (15iii)$$

$$\mu = \mu_2, \quad x > 0. \quad (15iv)$$

The parameters of the problem are μ_0, l, η, a, b . The basic viscosity is depending on a, b , and is continuous on Γ_1 , where the surface tension is zero.

Here μ depends only on x , but the perturbation μ' is function of (x, y, t) . The proof is as following. The problem (12)--(15) is linear. The first step is to consider the perturbation of basic velocity

$$u'(x, y, t) = f(x) \exp(i\alpha y + \sigma t). \quad (16)$$

where α is the wave number in direction y , orthogonal on the displacing direction Ox ; σ is the growth constant (in time) of the perturbations. From (12)--(14) we get

$$v' = -\frac{f_x}{i\alpha} \exp(i\alpha y + \sigma t), \quad p' = -\frac{\mu}{\alpha^2} f_x \exp(i\alpha y + \sigma t).$$

Cross differentiating the pressure we obtain

$$\begin{aligned} \frac{\partial}{\partial y} [\mu u' + \mu' U] &= \frac{\partial}{\partial x} [\mu v']; \\ \frac{\partial \mu'}{\partial y} &= \frac{1}{U} \left[-\frac{\mu_x}{i\alpha} f_x - \frac{\mu}{i\alpha} f_{xx} - \mu \cdot i\alpha \cdot f \right] \exp(i\alpha y + \sigma t). \end{aligned}$$

The above relations give us

$$\mu'(x, y, t) = h(x) \exp(i\alpha y + \sigma t); \quad h(x) = -\frac{1}{\alpha^2 U} \left\{ -(\mu f_x)_x + \alpha^2 \mu f \right\}, \quad (17)$$

therefore the second order partial derivatives of μ' are given by:

$$\frac{\partial^2 \mu'}{\partial x^2} = h(x)_{xx} \exp(i\alpha y + \sigma t); \quad \frac{\partial^2 \mu'}{\partial y^2} = -\alpha^2 h(x) \exp(i\alpha y + \sigma t).$$

We use (15i) and get the following *stability system*, where $x \in (-l, 0)$:

$$-(\mu f_x)_x + \mu \alpha^2 f = -\alpha^2 U h; \quad (18)$$

$$\eta h_{xx} - (\sigma + \eta \alpha^2) h = \alpha f. \quad (19)$$

In the following, we derive the boundary conditions for (18)--(19). The perturbed interfaces are described by the relations

$$x = g(0, y, t), \quad g_t = u', \quad g(0, y, t) = \frac{f(0)}{\sigma} \exp(i\alpha y + \sigma t);$$

$$x = -l + g(-l, y, t), \quad g_t = u', \quad g(-l, y, t) = \frac{f(-l)}{\sigma} \exp(i\alpha y + \sigma t),$$

therefore the limit values of the pressure on the interface $x=0$ can be approximated by:

$$p(0 + g, y, t) \approx P(0 + g, y, t) + p'(0, y, t) \approx P(0, y, t) + \frac{\partial P}{\partial x}(0, y, t) g(0, y, t) + p'(0, y, t),$$

$$p^+(0) \approx P(0) - \mu^+(0) U \frac{f(0)}{\sigma} \exp(i\alpha y + \sigma t) - \mu^+(0) \frac{1}{\alpha^2} f_x^+(0) \exp(i\alpha y + \sigma t),$$

$$p^-(0) \approx P(0) - \mu^-(0) U \frac{f(0)}{\sigma} \exp(i\alpha y + \sigma t) - \mu^-(0) \frac{1}{\alpha^2} f_x^-(0) \exp(i\alpha y + \sigma t).$$

Similar relations are obtained for limit values of the pressure on $x=-l$. We use the above relations to obtain the boundary conditions for stability system (18)--(19), as follows:

a) *Boundary conditions for f.* The viscosity is continuous on $x=-l$ and the surface tension is zero. On $x=0$ we have the viscosity jump $[\mu_2 - \mu(0)]$ and the surface tension T . On $x=-l$, $x=0$, we consider the first approximation of Laplace's law

$$p^+(0) - p^-(0) \approx T g_{yy}(0, y, t); \quad (20i)$$

$$p^+(-l) - p^-(-l) \approx 0. \quad (20ii)$$

We differentiate the pressure and get the equation of f out of the mixture:

$$f_{xx} - \alpha^2 f = 0.$$

The perturbations must be zero far from the interfaces (that means $u'=0$), therefore the above equation gives us the expression of f out of the mixture:

$$f(x) = e^{\alpha(x+l)} f(-l), \quad x < -l; \quad f(x) = e^{-\alpha x} f(0), \quad x > 0. \quad (21)$$

From here we obtain the following limit values of f :

$$f_x^-(-l) = \alpha f(-l), \quad f_x^+(0) = -\alpha f(0).$$

We use the above relations and from (20)--(21) we get

$$f_x^+(-l) = \alpha f(-l); \quad (22)$$

$$f_x^-(0) = \frac{1}{\mu(0)} \left\{ \frac{1}{\sigma} (U\alpha^2(\mu_2 - \mu(0)) - \alpha^4 T) - \mu_2 \alpha \right\} f(0) = (e\lambda + q) f(0); \quad (23)$$

$$e = \frac{1}{\mu(0)} \left\{ U\alpha^2(\mu_2 - \mu(0)) - \alpha^4 T \right\}; \quad \lambda = \frac{1}{\sigma}; \quad (24)$$

$$q = -\frac{\mu_2 \alpha}{\mu(0)}. \quad (25)$$

Here $\mu^+(-l)$, $f_x^+(-l)$ are the 'right' limits in $x=-l$, and $\mu^-(0)$, $f_x^-(0)$ are the 'left' limits in $x=0$

b) *Boundary conditions for h.* We use (11c) and get:

$$h(0) = h(-l) = 0 \quad (26)$$

5. THE APPROXIMATION PROCEDURE

We have to estimate the growth constant σ appearing in (18)--(19) with boundary conditions (22)--(26). The system (18)--(19) can be reduced to a single fourth order equation for the eigenfunction f . But, as we can see above, we have only two boundary conditions for f . In this situation, it is more useful to use two equations of second order, both with two boundary conditions, but coupled. In following sections we obtain an upper estimate for σ , by using this procedure. In this section we derive an approximated form of our stability system.

Consider $(M-1)$ points in the segment $[-l, 0]$: $x_M = -l < x_{M-1} < x_{M-2} < \dots < x_1 < x_0 = 0$ with the constant discretization step $d = x_i - x_{i+1}$. For the lateral derivatives appearing in (22)-(23) we use the approximations

$$(f_0 - f_1)/d = (e\lambda + q)f_0; \quad \left(\frac{1}{de} - \frac{q}{e} \right) f_0 - \frac{1}{de} f_1 = \lambda f_0; \quad (27)$$

$$(f_{M-1} - f_M)/d = \alpha f_M; \quad f_M = \frac{1}{1 + \alpha d} f_{M-1}; \quad (28)$$

with

$$e = \frac{U\alpha^2(\mu_2 - \mu(0)) - \alpha^4 T}{\mu(0)}, \quad q = \frac{\mu_2 \alpha}{\mu(0)}. \quad (29)$$

The values $f_0, f_1, f_2, \dots, f_{M-1}; h_1, h_2, \dots, h_{M-1}$ (the values of functions in the considered interior points) are unknown and we use the following approximations

$$h_x(y) = \frac{h(y+d/2) - h(y-d/2)}{d}; \quad h_{xx}(y) = \frac{h(y+d) - 2h(y) + h(y-d)}{d^2}.$$

Two kinds of indices will be used:

$$j, k = 0, 1, 2, \dots, (M-1); \quad i, m = 1, 2, \dots, (M-1),$$

then (27)--(29) give us the discretized form of our problem:

$$A_{00}f_0 + A_{01}f_1 = \lambda f_0; \quad (30)$$

$$A_{ik}f_k = -\alpha^2 U h_i; \quad (31)$$

$$\{\eta B_{im} - (\sigma + \alpha^2 \eta)\} h_m = a f_i, \quad (32)$$

where A and B are two *tridiagonal* matrices, given by the following expressions:

$$A_{00} = \left(\frac{1}{de} - \frac{q}{e} \right), \quad A_{01} = -\frac{1}{de}, \quad A_{0i} = 0, \quad \forall i > 1, \quad (33)$$

$$A_{i,i-1} = -\frac{\mu_{i-1/2}}{d^2}, \quad A_{i,i} = \mu_i \alpha^2 + \frac{\mu_{i-1/2} + \mu_{i+1/2}}{d^2}, \quad A_{i,i+1} = -\frac{\mu_{i+1/2}}{d^2}, \quad 1 \leq i \leq M-2, \quad (34)$$

$$A_{M-1, M-2} = -\frac{\mu_{n-1/2}}{d^2},$$

$$A_{M-1, M-1} = \left\{ \frac{\mu_{n-1/2} - \mu_{n+1/2}}{d^2} + \alpha^2 \mu_n - \frac{\mu_{n+1/2}}{d^2} \cdot \frac{1}{1+d\alpha} \right\}, \quad (35)$$

$$B_{ii} = -\frac{2}{d^2}, \quad B_{i-1,i} = B_{i,i+1} = \frac{1}{d^2}, \quad 1 \leq i \leq M-1. \quad (36)$$

We get only one equation for f , by using (31)-(32):

$$h_i = -\frac{a}{\sigma + \alpha^2 \eta} f_i + \frac{1}{\sigma + \alpha^2 \eta} \eta B_{im} h_m,$$

$$A_{ik} f_k = -\alpha^2 U h_i = (-\alpha^2 U) \left\{ -\frac{a}{\sigma + \alpha^2 \eta} f_i + \frac{1}{\sigma + \alpha^2 \eta} \eta B_{im} h_m \right\}.$$

From (31) we obtain $A_{mk} f_k = -\alpha^2 U h_m$. We replace h_m with the expression in *the last term* of the right hand side of the above formula. The final form of our discretized system is (recall $1 \leq i, m \leq M-1$ and $0 \leq k \leq M$):

$$A_{00}f_0 + A_{01}f_1 = \lambda f_0; \quad (37)$$

$$A_{ik} f_k = \frac{1}{\sigma + \alpha^2 \eta} (a \alpha^2 U) f_i + \frac{1}{\sigma + \alpha^2 \eta} \eta B_{im} A_{mk} f_k. \quad (38)$$

The above system contains only the eigenfunction f . Then the used approximation is useful for avoiding a fourth order differential equation for f , which appears in the exact form. There are $(M-1)$ equations and M unknowns f_k . The product BA is appearing, therefore the solution is not obvious. The difficulty is given by the matrix A appearing in (38). This matrix is not quadratic, therefore not invertible.

The growth constant σ and the eigenvalues λ are complex numbers. We use the notation $\sigma = \sigma_1 + i\sigma_2$, $\lambda = \lambda_1 + i\lambda_2$; $\sigma_i, \lambda_i \in \mathbf{R}$; $\lambda = 1/\sigma$.

In the following, we give an upper estimate of the real part of the growth constant, denoted by σ_1 . Consider the relation (38) in the equivalent form

$$(\sigma + \alpha^2 \eta) \sum_{k=0}^{M-1} A_{ik} f_k = (a\alpha^2 U) f_i + \eta \sum_{m=1}^{M-1} B_{im} \sum_{k=0}^{M-1} A_{mk} f_k; \quad i = 1, \dots, M-1. \quad (39)$$

We use the following notation

$$y_i = \sum_{k=0}^{M-1} A_{ik} f_k, \quad (40)$$

then (39) becomes

$$(\sigma + \alpha^2 \eta) y_i = a\alpha^2 U f_i + \eta \sum_{m=1}^{M-1} B_{im} y_m. \quad (41)$$

Let $VM = \max\{|y_i|, i = 1, 2, \dots, M-1\}$. Then we have the following possible relations

$$VM = |y_1| \Rightarrow |\sigma + \alpha^2 \eta - \eta B_{11}| \leq a\alpha^2 U \frac{|f_1|}{|y_1|} + \eta |B_{12}| \quad (42)$$

or

$$VM = |y_s|, \quad 1 < s < M-1 \Rightarrow |\sigma + \alpha^2 \eta - \eta B_{ss}| \leq a\alpha^2 U \frac{|f_s|}{|y_s|} + \eta (|B_{s,s-1}| + |B_{s,s+1}|) \quad (43)$$

or

$$VM = |y_n|, \quad n = M-1 \Rightarrow |\sigma + \alpha^2 \eta - \eta B_{n,n}| \leq a\alpha^2 U \frac{|f_n|}{|y_n|} + \eta |B_{n,n-1}|. \quad (44)$$

Here α, B, η are real, then we have the inequality

$$\sigma_1 + \alpha^2 \eta - \eta B_{j,j} \leq |\sigma + \alpha^2 \eta - \eta B_{j,j}|.$$

In the above equations (42)--(44) we use the obvious relations

$$B_{11} + |B_{12}| = -2/d^2 + 1/d^2 \leq 0; \quad B_{ss} + |B_{s,s-1}| + |B_{s,s+1}| = -2/d^2 + 1/d^2 + 1/d^2 = 0,$$

$$B_{M-1,M-1} + |B_{M-1,M-2}| = -2/d^2 + 1/d^2 \leq 0.$$

The last relation and the estimates (42)--(44) give us the final upper bound for σ_1 :

$$\sigma_1 + \alpha^2 \eta \leq a\alpha^2 U \frac{|f_s|}{|y_s|}, \quad |y_s| = \max\{|y_i|, 1 \leq i \leq M-1\}, \quad (45)$$

where y_i are given by the definition (40).

Remark 1. The procedure used in this section is quite similar with the Gerschgorin's localization result for the eigenvalues of the matrix equation $Ax = \lambda x$. However, in our case, the new term $|f_i/y_i|$ is appearing

Remark 2. The estimate (45) allows us the following conclusion: a large enough diffusion coefficient can improve the flow stability. This means that the growth constant becomes negative for large diffusion coefficient η .

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