

The Onset of Taylor-Görtler Vortices in Impulsively Decelerating Swirl Flow

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Abstract—The onset of hydrodynamical instability induced by impulsive spin-down to rest in a cylinder containing a Newtonian fluid is analyzed by using propagation theory. It is well-known that the primary transient swirl flow is laminar, but with initial high velocities secondary motion sets in at a certain time. The dimensionless critical time τ_c to mark the onset of instability is presented here as a function of the Reynolds number Re . Available experimental data indicate that for large Re deviation of the velocity profiles from their momentum diffusion occurs starting from a certain time $\tau \approx 4\tau_c$. This means that secondary motion is detected at this characteristic time. It seems evident that during $\tau_c \leq \tau \leq 4\tau_c$, secondary motion is relatively very weak and the primary diffusive momentum transfer is dominant.

Key words: Taylor-Görtler Vortex, Swirl Flow, Reynolds Number, Hydrodynamical Instability, Propagation Theory

INTRODUCTION

It is well-known that in the primary laminar flows along concavely curved walls, the destabilizing action of the centrifugal forces can produce secondary motion in the form of vortices. The related hydrodynamical instabilities usually lead to Taylor vortices in the flow between concentric cylinders or Görtler ones in the boundary layer flow. The instability problem of transient laminar swirl flow in a cylinder is closely related to that of Taylor-Görtler vortices. The onset of instability caused by spin-down, when a rotating liquid-filled cylinder is suddenly brought to rest, was first investigated experimentally by Euteneuer [1972]. The initial laminar flow evolves into a secondary flow pattern which consists of a series of Taylor-like vortices. This kind of secondary flow plays an important role in mixing in a vertical Bridgman crystal growth system where the crucible is rotated to improve mixing [Yeckel and Derby, 2000]. In this transient boundary-layer system the critical time τ_c to mark the onset of secondary motion becomes an important question.

A related instability analysis has been conducted by using the energy method [Neitzel and Davis, 1980; Neitzel, 1982a] and also by employing direct numerical simulation [Neitzel and Davis, 1981]. A similar stability problem, where a fluid is filled between the two concentric cylinders and the rotation of the inner cylinder is impulsively started from rest, i.e., the spin-up problem, has been analyzed by the amplification theory [Chen and Kirchner, 1971], the frozen-time model [Chen and Kirchner, 1971], the energy method [Neitzel, 1982b] and the maximum-Taylor-number criterion [Tan and Thorpe, 2003]. The amplification theory model requires the initial conditions and the criterion to define detection of manifest convection. The frozen-time model is based on linear theory and yields the critical time as the parameter. The energy method suggests lower bounds on the experimental onset times. The amplification theory and the energy method are quite popular, but they require a large number of tedious computations. Even though the max-

imum-Taylor-number criterion is the simplest one, it seems to lack physical insights. These models take advantage of the similarity between Taylor instability and Rayleigh-Bénard instability.

Another model to analyze time-dependent convective instability problems is propagation theory [Choi et al., 1998; Kim et al., 2002], which deals with thermal instability problems of developing, non-linear temperature profiles in rapidly heated systems. In propagation theory, any kind of arbitrariness such as the initial conditions and the criterion to define detection of manifest convection and severe calculation burden is not required. Therefore, propagation theory can be said to be a deterministic and relatively simple method. This model assumes that at $t=t_c$ infinitesimal temperature disturbances are propagated mainly within the thermal penetration depth Δ_T and with this length scaling factor all the variables and parameters having the length scale are rescaled. In a usual deep-pool conduction system of $\Delta_T \propto \sqrt{\alpha t}$, the most important parameter becomes the time-dependent Rayleigh number, which is yielded by replacing the length scale in the Rayleigh number with Δ_T . Here α is the thermal diffusivity. The resulting stability criteria have compared well with experimental data of various systems such as solidification [Hwang and Choi, 1996], Marangoni-Benard convection [Kang and Choi, 1997; Kang et al., 2000] and Benard convection in porous media [Yoon and Choi, 1989].

Here we will extend the propagation theory, which has been employed to analyze time-dependent diffusive problems, to the hydrodynamical instability induced by an aforementioned impulsively-stopped swirl flow. The resulting predictions will be discussed in comparison with available experimental results.

THEORETICAL ANALYSIS

1. Governing Equations

The system considered here is a Newtonian fluid confined in a cylinder of radius R . Let the axis of the inner cylinder be along the vertical z' -axis under the cylindrical coordinates (r', θ, z') and the corresponding velocities be U, V and W . The entire fluid/cylinder system is assumed to be in an initial state of rigid-body rotation with

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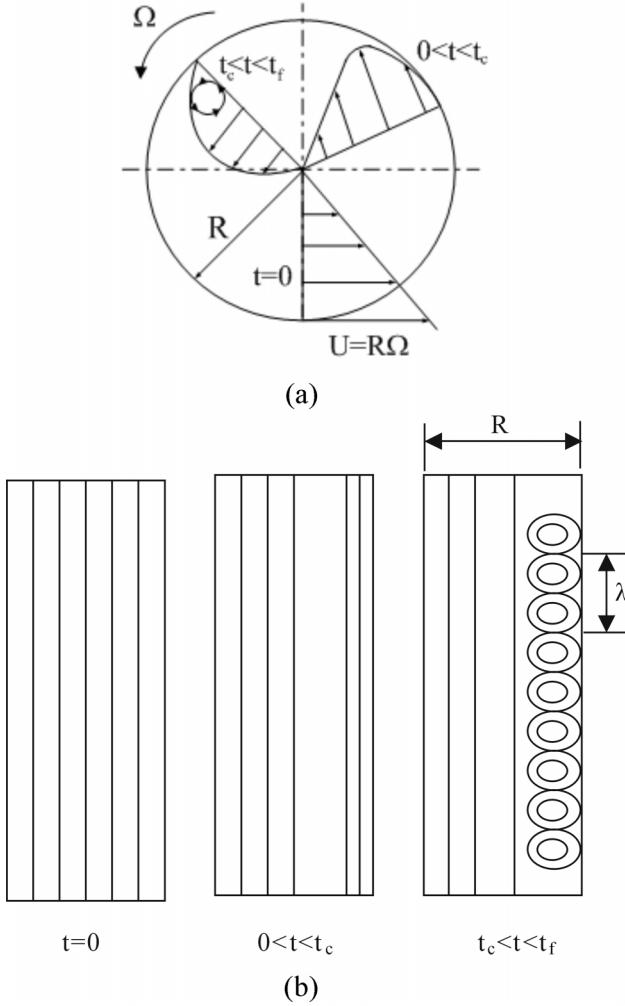


Fig. 1. Schematic views of the basic system considered here: (a) top view and (b) streamlines.

a constant angular velocity Ω . Starting from time $t=0$, the cylinder is impulsively stopped. The ensuing unsteady swirl flow shows the state of spin-down. A schematic diagram of the present system is shown in Fig. 1. Due to the asymptotically unconditionally-stable characteristic of this flow the secondary motion disappears after t_c . Such swirl flow encounters instabilities in the form of Taylor-Görtler vortices and the governing equations of the flow field are expressed as

$$\nabla \cdot \mathbf{U} = 0, \quad (1)$$

$$\left\{ \frac{\partial}{\partial t} + \mathbf{U} \cdot \nabla \right\} \mathbf{U} = -\frac{1}{\rho} \nabla P + \nu \nabla^2 \mathbf{U}, \quad (2)$$

where \mathbf{U} , P , ν and ρ represent the velocity vector, the dynamic pressure, the kinematic viscosity and the density, respectively.

For the case of constant physical properties the primary-velocity field is represented:

$$\frac{\partial V_0}{\partial t} = \nu D' D'' V_0, \quad (3)$$

with the following initial and boundary conditions,

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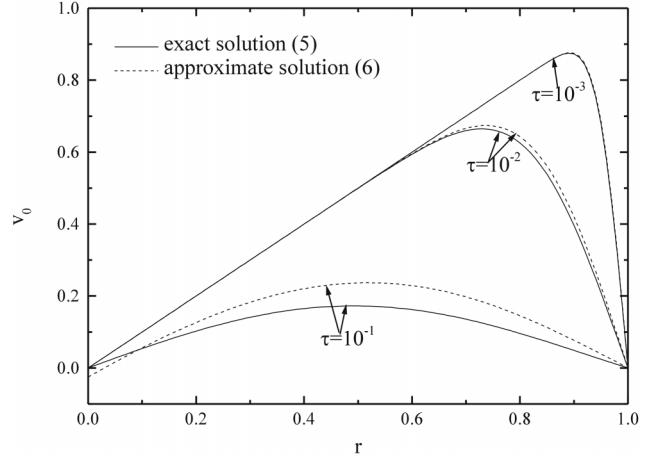


Fig. 2. Primary-velocity profiles.

$$V_0(0, r) = r\Omega, V_0(t, 0) = 0, V_0(t, R) = 0. \quad (4a, b, c)$$

where $D' = \partial/\partial r'$ and $D'' = D' + 1/r'$. Neitzel [1982a] obtained the exact solution as

$$V_0(\tau, r) = \frac{V_0}{R\Omega} = -2 \sum_{i=1}^{\infty} \frac{J_1(\beta_i r)}{\beta_i J_0(\beta_i)} \exp(-\beta_i^2 \tau), \quad (5a)$$

where β_i are the roots of

$$J_1(\beta_i) = 0, \quad (5b)$$

where J_i denotes Bessel functions of order i of the first kind. For small time the velocity approaches the following complementary error function:

$$v_0 = 1 - y - \text{erfc} \left[\frac{y}{\sqrt{4\tau}} \right], \quad (6)$$

where $v_0 = V_0/(R\Omega)$, $r = r'/R$, $y = (1-r)$, and $\tau = vt/R^2$. The instantaneous base flow profile is shown in Fig. 2. For $\tau \leq 10^{-3}$, Eq. (6) approximates the exact solution (5) very well. Since the present study concerns the deep-pool system of small time, Eq. (6) is used in the stability analysis. The problem is to find the dimensionless critical time τ_c to mark the onset of instability, which grows with time.

2. Stability Equations

The typical disturbances which are observed experimentally are well represented by

$$(U_1, V_1, P_1) = (u', v', p') \cos kz', \quad (7a)$$

$$W_1 = w' \sin kz', \quad (7b)$$

where k is the wavenumber and the primed quantities representing disturbance amplitudes are a function of r' and t . The two-dimensional perturbed quantities are periodic in the z' -direction. Under linear theory the stability equations of amplitude functions are obtained when w' and p' are eliminated. Under the deep-pool approximation of small τ , where $\partial/\partial r + 1/r \approx \partial/\partial r$, the resulting dimensionless amplitude equations are represented by

$$\left(D'^2 - k^2 - \frac{1}{\nu \partial t} \right) (D'^2 - k^2) u' = 2 \frac{V_0 k^2}{r' \nu} v', \quad (8)$$

$$\left(D'^2 - k^2 - \frac{1}{\nu \partial t} \right) v' = \frac{D' V_0}{\nu} u', \quad (9)$$

and the no-slip boundary conditions at $r=R$ are

$$u'=D'u'=v'=0 \quad \text{at} \quad r=R, \quad (10a)$$

The requirement that all the velocity components be bound at $r=0$ results in

$$u'=DD'u'=v'=0 \quad \text{at} \quad r=0 \quad (10b)$$

The derivation of all the above equations is described in detail by Chandrasekhar [1961] and Neitzel [1982a].

The propagation theory employed to find the onset time of instability, i.e., the critical time τ_c , requires the assumption that in deep-pool systems of small time, the perturbed angular velocity component v' is propagated mainly within the hydrodynamic boundary-layer thickness $\Delta(\propto \sqrt{v\tau})$ at the onset of instability. The resulting scale relations for perturbed quantities from Eq. (2) are given by

$$v' \sim \frac{V_0}{\Delta^2} v', \quad (11)$$

$$u' \sim \frac{V_0}{\Delta} v' \sim \frac{v'}{\Delta^2}, \quad (12)$$

from the balances among acting forces (viscous, centrifugal, and inertial). Now, the following relation between dimensionless amplitudes of u' and v' is obtained in dimensionless form:

$$\frac{u}{v} \sim \delta^2 \sim \tau, \quad (13)$$

where $\delta(\equiv \Delta/R \propto \sqrt{\tau})$ is the usual dimensionless boundary-layer thickness. Here $u=uv/(R^3\Omega^2)$ and $v=2v/(R\Omega)$. They are the nondimensional velocity perturbations, which were used by Chandrasekhar [1961], and Chen and Kirchner [1971]. Now, the following relation is produced from the above relations:

$$Ta^* u^* (\partial v^* / \partial r) \sim v, \quad (14)$$

where $Ta^* = \tau^{3/2} Re^2$ and $u^* = u/\tau$. Here Re and Ta^* are the Reynolds number and the Taylor number (or Görtler number), based on the boundary-layer thickness, respectively:

$$Re = \frac{\Omega R^2}{v}, \quad Ta^* = \left(\frac{V_0 \Delta}{v} \right)^2 \frac{R}{\Delta}. \quad (15)$$

where Ta^* is the Taylor number based on the Rayleigh thickness ($\sqrt{v\tau}$). This has been used in the spin-up problem [Otto, 1993].

Now, for small time we introduce the similarity variable $\zeta = y/\tau^{1/2}$ and assume that dimensionless amplitude functions of disturbances have the forms of

$$[u(\tau, \zeta), v(\tau, \zeta)] = [\tau^{n+1} u^*(\zeta), \tau^n v^*(\zeta)], \quad (16)$$

which satisfies the above relations. We set $n=0$. This means that the amplitude function v is a function of ζ only by following the behavior of v_0 for small τ , as shown in Eq. (6). The case of $n<0$ is not rational since $v^* \rightarrow \infty$ as $\tau \rightarrow 0$. For $n \geq 0$, the case of $n=0$ yields the fastest growing disturbances, i.e., the minimum Reynolds number. A similar treatment can be found in problems of transient Bénard-type convection [Choi et al., 1998; Kang et al., 2000]. Furthermore, the relation of $Ta^* \equiv \text{constant}$ for large Re is shown even in theoretical results from the energy method [Neitzel, 1982a].

By the above reasoning we set $u=u^*(\zeta)$ and $v=v^*(\zeta)$. For bound-

ary-layer flow systems of $\delta \propto \sqrt{\tau}$, the dimensionless time τ plays dual roles of time and boundary-layer thickness. Now, the self-similar stability equations are obtained in dimensionless form from Eqs. (12) and (13) as

$$\left[(D^2 - a^{*2})^2 + \frac{1}{2} (\zeta D^3 - a^{*2} \zeta D + 2a^{*2}) \right] u^* = \frac{V_0}{1-y} a^{*2} v^*, \quad (17)$$

$$(D^2 - a^{*2} + \frac{1}{2} \zeta D) v^* = 2Ta^* (D^* v_0) u^*, \quad (18)$$

where $D=d/d\zeta$, $y=\zeta\sqrt{\tau}$ and $a^* = a\sqrt{\tau}$. Here Ta^* and a^* have been treated as eigenvalues and a is the dimensionless wavenumber ($=kR$) in the z' -direction. The proper boundary conditions are

$$u^* = Du^* = v^* = 0 \quad \text{at} \quad \zeta = 0, \quad (19a)$$

$$u^* = D^2 u^* = v^* = 0 \quad \text{as} \quad \zeta \rightarrow \infty. \quad (19b)$$

Now, the minimum value of Ta^* should be found in the plot of Ta^* vs. a^* under the principle of the exchange of stabilities. In other words, the minimum value of τ , i.e., τ_c , and its corresponding wavenumber, a_c , should be obtained for a given Re . Since time has been frozen by letting $\partial(\cdot)/\partial r=0$ under the frame of coordinates τ and ζ instead of τ and y , the propagation theory may be called the relaxed frozen-time model by treating τ as the parameter, but it involves the time dependency implicitly.

3. Solution Procedure

To find eigenvalues and eigenfunctions for differential equations, several methods such as compound matrix method and shooting method are proposed [Straughan, 1992]. In the present study the stability Eqs. (17)-(19) are solved by employing the latter method. In order to integrate these stability equations the proper values of $D^2 u^*$, $D^3 u^*$ and Dv^* at $\zeta=0$ are assumed for a given a^* . Since the stability equations and their boundary conditions are all homogeneous, the value of $D^2 u^*(0)$ can be assigned arbitrarily and the value of the parameter Ta^* is assumed. This procedure can be understood easily by taking into account the characteristics of eigenvalue problems [Straughan, 1992]. After all the values at $\zeta=0$ are provided, this eigenvalue problem can proceed numerically.

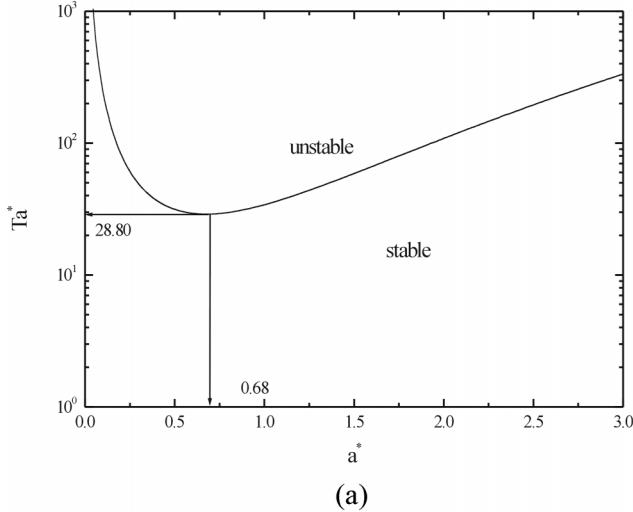
Integration is performed from $\zeta=0$ to a fictitious upper boundary with the fourth order Runge-Kutta-Gill method. If the guessed values of Ta^* , $D^3 u^*(0)$ and $Dv^*(0)$ are correct, u^* , $D^2 u^*$ and v^* will vanish at the axis of rotation. Since disturbances decay exponentially outside the boundary-layer thickness, the incremental change of Ta^* also decays fast with increasing a fictitious outer boundary thickness. This behavior enables us to extrapolate the eigenvalue to the axis of rotation. A typical stability curve is shown in Fig. 3(a) and the minimum Ta^* -value is found to be 28.80 with its corresponding a^* value of 0.68.

RESULTS AND DISCUSSION

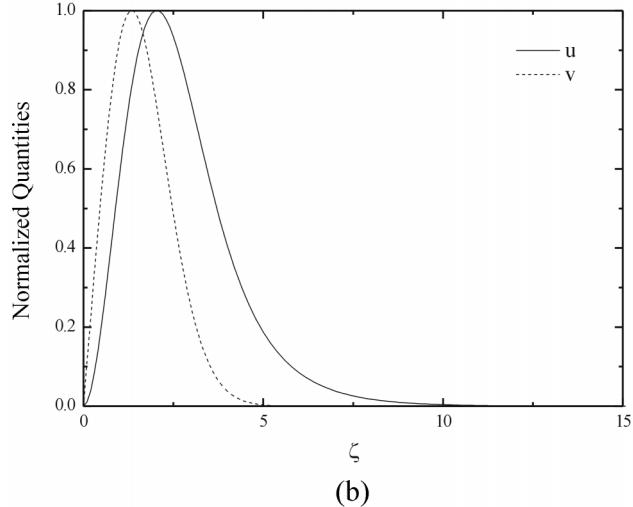
For the limiting case of $\tau \rightarrow 0$, the stability criteria under the single mode of instability have been obtained from the propagation theory. The critical conditions from Fig. 3(a) can be converted into

$$\tau_c = 9.40 Re^{-4/3} \quad \text{and} \quad a_c = 0.22 Re^{2/3} \quad \text{as} \quad \tau \rightarrow 0. \quad (20)$$

At this critical condition the profiles of amplitude functions are featured in Fig. 3(b). The critical time τ_c to mark the onset of a fastest



(a)



(b)

Fig. 3. Instability conditions for small time of $\tau_c \rightarrow 0$ from the propagation theory: (a) marginal stability curve and (b) amplitude profiles at $\tau = \tau_c$.

growing instability decreases with increasing Re . It is known that disturbances of the angular velocity are confined mainly within the hydrodynamic boundary layer of the primary flow (see Fig. 2). The same trend is also shown in Rayleigh-Bénard problems [Yang and Choi, 2002; Kim et al., 2002].

Now, the above results are compared with the available experimental data [Euteneuer, 1972] and also predictions. Experimentally, secondary motion was observed at $t = t_0$. Neitzel and Davis [1980] and Neitzel [1982a] employed the energy method, where the time evolution of the volume-integrated kinetic energy of disturbances for a given wavelength was monitored. They suggested a strong stability limit t_s , up to which the kinetic energy of a most dangerous mode of disturbances should decay and a marginal stability limit t_m , from which the kinetic energy exceeds the assumed, initial kinetic energy. Starting from $t = t_s$, the kinetic energy increases with time. Their concept of stability limit is well summarized in Fig. 1 of Neitzel [1982a]. The corresponding dimensionless times, τ_m and τ_s , are compared with τ_c and τ_o in Fig. 4. For transient instability problems on thermal convection, Foster [1969] commented that with correct dimensional relations the relation of $\tau_o \approx 4\tau_c$ would be kept for the case

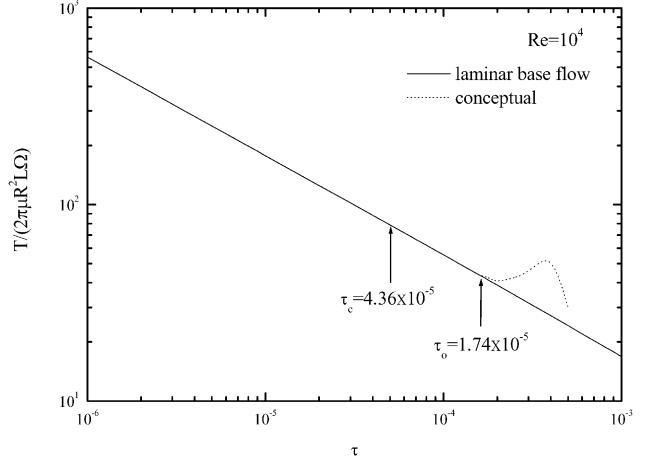


Fig. 4. Typical conceptual diagram of temporal behavior of torque.

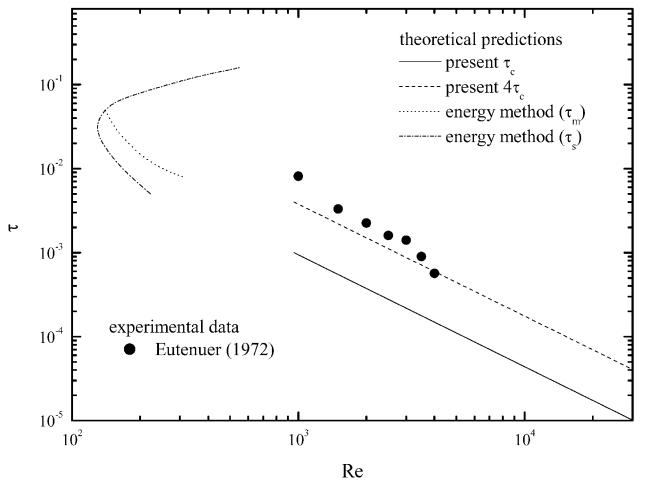


Fig. 5. Comparison of predictions of characteristic times with available experimental data.

of rapid heating in horizontal fluid layers. This relation has been shown in predictions obtained from the propagation theory for large-Prandtl systems [see the work of Choi et al., 1998]. For the spin-up system, based on the amplification theory, Chen and Kirchner [1971] reported a similar trend. Their results show that the characteristic time τ_c at which disturbances first tend to grow is about one-fourth of the time τ_o at which convective motion is clearly observable experimentally. All the above-mentioned models imply that a growth period for disturbances to grow is required until they are detected experimentally. Therefore, it seems evident that the predicted onset time τ_c is smaller than the detection τ_o . This means that a fastest growing mode of instabilities, which set in at $t = t_c$, will grow with time until manifest motion is first detected experimentally. This conceptual behavior of torque is illustrated in Fig. 4. The condition (20) yields $\tau_c = 4.36 \times 10^{-5}$ for $Re = 10^4$ as shown in Fig. 4. Linear theory is applied up to $\tau \approx \tau_o$ from which the flow profile deviates from base flow and therefore, manifest convection can be observed at $\tau = \tau_o \approx 4\tau_c$. The present predictions are consistent with the experimental data for $Re > 1,000$ as shown in Fig. 5 where we infer upper bounds of τ_o by noting the earliest time for which wavelength data are presented in Fig. 3 of Euteneuer's [1972]. In Fig. 6 the predicted cri-

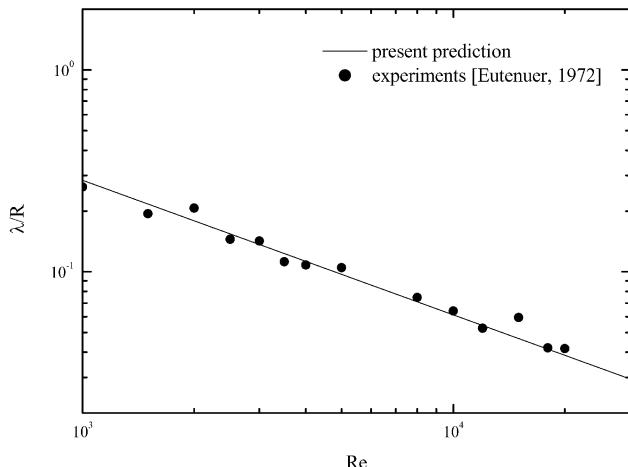


Fig. 6. Comparison of the dimensionless critical wavenumber a with respect to the Reynolds number Re .

tical wavelength ($\lambda_c=2\pi R/a_c$) is compared with the experimental ones given in Fig. 4 of Euterneuer [1972]. It seems evident that the cell size is almost constant during $t_c \leq t \leq t_v (\approx 4t_c)$ but its radial growth will be continued. This viewpoint is also seen in the results of the energy method.

It is known that the propagation theory, which is based on self-similar transformation and normal mode analysis, is a powerful method to predict the stability criteria reasonably well in simple systems, hydrodynamic or thermal. But its validity should be justified in the future by employing the numerical method to take the full nonlinear effects in Eq. (2) into the instability analysis like Choi et al.'s [2003] work on thermal convection.

CONCLUSION

The onset of a fastest growing, axisymmetric instability in transient swirl flow has been investigated by the propagation theory which was originally devised for the problems in transient Benard-like convection. The present predictions of $\tau_c \equiv 4\tau_c$ to represent the characteristic time first detection of manifest axisymmetric flow compare well with available experimental data. It is interesting that the propagation theory yields instability criteria compatible with experimental observations in simple diffusive systems, hydrodynamical or thermal, which involve a similarity variable.

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NOMENCLATURE

| | |
|-----|--------------------------------------|
| a | : dimensionless wavenumber, kR |
| k | : wavenumber |
| L | : length of cylinder |
| P | : pressure |
| p | : dimensionless pressure disturbance |
| R | : radius of cylinder |

| | |
|--------------------|---|
| Re | : Reynolds number, $\Omega R^2/v$ |
| T | : torque |
| Ta^* | : Taylor number based on boundary-layer thickness, $\tau^{3/2}Re^2$ |
| (U, V, W) | : velocities in cylindrical coordinates |
| (u, v, w) | : dimensionless velocity disturbances in cylindrical coordinates |
| (u', v', w') | : velocity disturbance amplitudes in cylindrical coordinates |
| (r', θ, z') | : cylindrical coordinates |
| (r, θ, z) | : dimensionless cylindrical coordinates |

Greek Letters

| | |
|------------|---|
| α | : thermal diffusivity |
| Δ | : boundary-layer thickness |
| Δ_T | : thermal boundary-layer thickness |
| δ | : dimensionless boundary-layer thickness |
| ζ | : dimensionless similarity variable, $y/\tau^{1/2}$ |
| λ | : wavelength, $2\pi R/a$ |
| ν | : kinematic viscosity |
| τ | : dimensionless time |

Subscripts

| | |
|-----|---------------------------|
| i | : inlet conditions |
| 0 | : basic quantities |
| 1 | : perturbation quantities |
| c | : critical conditions |

Superscript

| | |
|-----|--------------------------|
| $*$ | : transformed quantities |
|-----|--------------------------|

REFERENCES

Chandrasekhar, S., "Hydrodynamic and Hydromagnetic Stability," Oxford University Press, Oxford (1961).

Chen, C. F. and Kirchner, R. P., "Stability of Time-Dependent Rotational Couette Flow. Part 2. Stability Analysis," *J. Fluid Mech.*, **48**, 365 (1971).

Choi, C. K., Park, J. H., Park, H. K., Cho, H. J., Chung, T. J. and Kim, M. C., "Temporal Evolution of Thermal Convection in an Initially, Stably Stratified Fluid," Proc. Int. Symp. Transient Convective Heat and Mass Transfer in Single and Two-Phase Flows (Extended Abstracts), Cesme, pp. 63-66 (2003).

Choi, C. K., Kang, K. H. and Kim, M. C., "Convective Instabilities and Transport Properties in Horizontal Fluid Layers," *Korean J. Chem. Eng.*, **15**, 192 (1998).

Euteneuer, G.-A., "Die Entwicklung von Langswirbeln in Zeitlich Anwachsenden Grenzschichten an Konkaven Wänden," *Acta Mechanica*, **13**, 215 (1972).

Foster, T. D., "Onset of Manifest Convection in a Layer of Fluid with a Time-Dependent Surface Temperature," *Phys. Fluids*, **12**, 2482 (1969).

Hwang, I. G. and Choi, C. K., "An Analysis of the Onset of Compositional Convection in a Binary Melt Solidified from Below," *J. Crystal Growth*, **162**, 182 (1996).

Kang, K. H. and Choi, C. K., "A Theoretical Analysis of the Onset of Surface-tension-driven Convection in a Horizontal Liquid Layer Cooled Suddenly from Above," *Phys. Fluids*, **9**, 7 (1997).

Kang, K. H., Choi, C. K. and Hwang, I. G., "Onset of Marangoni Convection in a Suddenly Desorbing Liquid Layer," *AIChE J.*, **46**, 15 (2000).

Kim, M. C., Park, H. K. and Choi, C. K., "Stability of an Initially, Stably Stratified Fluid Subjected to a Step Change in Temperature," *Theoret. Comput. Fluid Dynamics*, **16**, 49 (2002).

Neitzel, G. P., "Marginal Stability of Impulsively Initiated Couette and Spin Decay," *Phys. Fluids*, **25**, 226 (1982a).

Neitzel, G. P., "Stability of Circular Couette Flow with Variable Inner Cylinder Speed," *J. Fluid Mech.*, **123**, 43 (1982b).

Neitzel, G. P. and Davis, S. H., "Energy Stability Theory of Decelerating Swirl Flows," *Phys. Fluids*, **23**, 432 (1980).

Neitzel, G. P. and Davis, S. H., "Centrifugal Instabilities during Spin-Down to Rest in Finite Cylinders. Numerical Experiments," *J. Fluid Mech.*, **102**, 329 (1981).

Otto, S. R., "Stability of the Flow Around a Cylinder: The Spin-up Problem," *IMA J. Appl. Math.*, **51**, 13 (1993).

Straughan, B., "The Energy Method, Stability, and Nonlinear Convection," Springer-Verlag, N.Y. (1992).

Tan, K.-K. and Thorpe, R. B., "Transient Instability of Flow Induced by an Impulsively Started Rotating Cylinder," *Chem. Eng. Sci.*, **58**, 149 (2003).

Yang, D. J. and Choi, C. K., "The Onset of Thermal Convection in a Horizontal Fluid Layer Heated from Below with Time-Dependent Heat Flux," *Phys. Fluids*, **14**, 930 (2002).

Yeckel, A. and Derby, J. J., "Effect of Accelerated Crucible Rotating on Melt Composition in High Pressure Vertical Bridgman Growth of Cadmium Zinc Telluride," *J. Crystal Growth*, **209**, 734 (2000).

Yoon, D.-Y. and Choi, C. K., "Thermal Convection in a Saturated Porous Medium Subjected to Isothermal Heating," *Korean J. Chem. Eng.*, **6**, 144 (1989).