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Publication date
2022

Document Version
Final published version

Citation (APA)
Bugeat, B., Boldini, P. C., & Pecnik, R. (2022). *On the new unstable mode in the boundary layer flow of supercritical fluids*. Paper presented at 12th International Symposium on Turbulence and Shear Flow Phenomena, TSFP 2022, Osaka, Virtual, Japan.

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ON THE NEW UNSTABLE MODE IN THE BOUNDARY LAYER FLOW OF SUPERCRITICAL FLUIDS

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ABSTRACT

Ren *et al.* (2019) recently studied the stability of the boundary layer flow over a flat plate for supercritical CO2. While only one unstable mode usually exists for boundary layer flows, the authors found an additional unstable mode, whose origin has so far not been identified. In the present work, we carry out a stability analysis in the general case of a fluid following the Van der Waals equation of state and flowing over a heated flat plate in the limit of zero Eckert number. In this framework, the second unstable mode is also recovered, ruling out an acoustic origin. From the Rayleigh equation derived in the presence of density gradients, a generalised inflection point (GIP) criterion of instability exists, similar to that of fully compressible flows. Inviscid stability calculations confirm the existence of an unstable mode in the presence of a GIP, which is linked to the additional second mode found at finite Reynolds numbers. A theoretical analysis is then carried out by approximating the momentum equation for a base flow exhibiting strong gradients of dynamic viscosity. It is shown that the origin of the GIP, and hence the additional unstable mode, is associated with a minimum of kinematic viscosity at the Widom line. The universality of this result beyond supercritical fluids is eventually discussed.

CONTEXT AND OBJECTIVES

Above certain conditions of temperature and pressure, fluids reach a supercritical state in which the distinction between liquid and gas states no longer exists. These fluids have important applications in industrial processes, in particular in power cycles (Iverson *et al.* (2013)) for which accurate models of heat transfers are required. Heat transfers depend dramatically on the flow regime; yet, little is known about the transition to turbulence in supercritical fluids. Ren *et al.* (2019) recently studied the stability of the boundary layer over a flat plate for supercritical CO2. The authors discovered a new linearly unstable mode in addition of the usual Tollmien-Schlichting wave. This mode was furthermore different from the acoustic unstable Mack's modes observed in compressible boundary layers. In their study, the authors considered temperature gradients generated by viscous heating. The new mode was then detected when sufficient viscous heating, controlled by the Eckert number, made the temperature profile cross the Widom line. This line, in the supercritical region of the thermodynamic space, divides the gas-like from the liquid-like behaviour of a fluid, and is then also called pseudo-boiling line (Banuti, 2015). It can be seen as an extension of the coexistence line between liquid and gas but greatly differs from it as thermodynamic properties continuously vary across it. Still, steep variations of the supercritical fluid properties are observed around the Widom line, in particular the density and the viscosity.

The aim of the present paper is to shed light on the origin and the nature of the new unstable mode by understanding the role played by the Widom line. A linear stability analysis using the Navier-Stokes equation is first carried out on a Van der Waals fluids, allowing us to recover the qualitative behaviour of Ren *et al.* (2019). Inviscid stability calculations are then performed in order to show the inflexional nature of this instability. Subsequently, we theoretically show that a generalised inflexion point (GIP) can appear when the temperature profile crosses the Widom line is crossed, and that it is fundamentally linked to the presence of a minimum of kinematic viscosity. A discussion on the universality of this instability, beyond supercritical fluids, is then opened.

APPROACH

The scope of this study is limited to a supercritical fluid flowing over a flat plate with an Eckert number set to zero (equivalently, the Mach number is also zero). This allows us to ignore acoustic effects, which were taken into account by Ren *et al.* (2019). The physical interpretation of the results is then simplified while the equations are less cumbersome. To

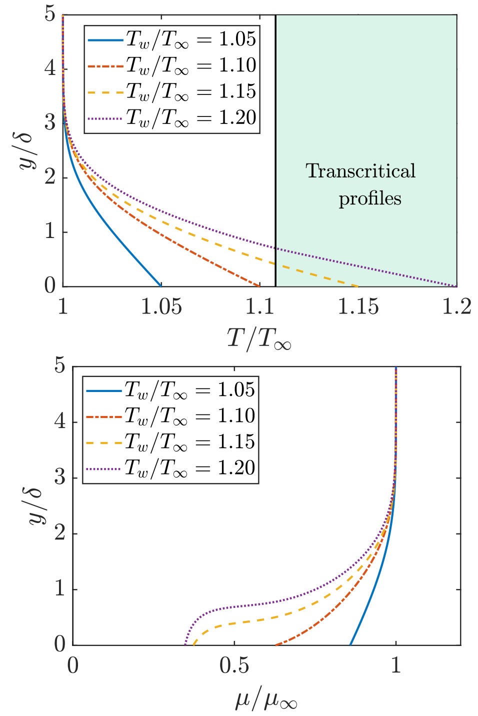

Figure 1: Base flow profiles for different wall temperatures. Left: temperature. Profiles that crosses the pseudo-boiling temperature, indicated by the vertical black line, are called transcritical. Right: dynamic viscosity.

trigger non-ideal effects in the absence of viscous heating generated by finite Eckert numbers, the flat plate is maintained at a prescribed temperature T_w larger than the freestream temperature T_∞. The Van der Waals equation of state is used along with the dynamic viscosity and thermal conductivity laws for supercritical fluids proposed by Jossi *et al.* (1962) and Stiel & Thodos (1964), respectively. The reduced pressure being fixed at $p/p_c = 1.08$, the pseudo-boiling temperature is found to be equal to $T_{pb}/T_\infty = 1.11$. Two wall temperatures below and two above T_{pb} are considered (figure 1). In the two latter cases, the temperature profile then crosses the Widom line: there is a wall-normal location at which the base flow verifies $\bar{T} = T_{pb}$. These cases are referred to as *transcritical*.

The Navier-Stokes equations are solved numerically in the limit of the boundary layer approximation in order to obtain steady, self-similar base flows for different wall temperatures (as just shown in figure 1). A local stability analysis is then carried out by decomposing small perturbations into normal modes. This involves linearising the Navier-Stokes equations written in the limit of zero-Eckert number. In this framework, density may only vary due to the effect of temperature, while the pressure is purely of hydrodynamic nature. An eigenvalue problem is eventually solved using a pseudo-spectral method (Orszag, 1971), from which the spatial growth rate, noted $-\alpha_i$, of each mode is obtained.

An inviscid linear stability solver is also used in this work in order to study the stability of the flow in the limit of infinitely large Reynolds numbers. It is based on the Rayleigh equation extended to a varying-density base flow. Following Boyd (1985), a parabolic complex-mapping is used to avoid the well-known critical layer singularity occurring for neutral modes. This allows us to accurately calculate small values of the growth rate.

VISCOUS STABILITY RESULTS

The results of the linear stability analysis are shown in figure 2, in which neutral curves are plotted in the Reynolds number-frequency space (Re, ω). When the wall temperature is below the pseudo-boiling temperature, one and only one unstable mode, termed mode A, is found. It is the Tollmien-Schlichting wave reminiscent of the incompressible case in the absence of temperature gradient. When T_w is increased and the Widom line is crossed, a new mode, termed mode B, appears in addition to mode A. It can be seen that it results from the destabilisation of a distinct eigenvalue from mode A since the two modes can be unstable at the same values of Re and ω. Mode B is further destabilised as T_w is increased, with the critical Reynolds number of instability dramatically decreasing and the magnitude of the growth rate increasing.

These results are qualitatively similar to those of Ren *et al.* (2019). This shows that mode B is not associated with acoustic effects since $Ec = 0$ in the present calculation. Moreover, while the authors studied supercritical CO_2 with non-analytical thermodynamic functions obtained from look-up tables, we here see that the simple Van der Waals equation of states associated with analytical diffusion laws produces the same observations. This indicates some degree of robustness of this new mode and shows that this simpler and more universal model contains the physical ingredients responsible for its growth, offering a framework that may be more suited for a fundamental analysis of this instability.

INVISCID STABILITY ANALYSIS

A linear inviscid stability analysis is carried out to determine the nature of mode B. When the wall temperature is below the pseudo-boiling temperature ($T_w/T_{pb} < 1$), no unstable mode is found. However, as soon as $T_w/T_{pb} > 1$, positive growth rate are detected (figure 3). In addition to the evidence of the inflexional nature of mode B, this result represents a new strong indication that this mode is intrinsically linked to the crossing the Widom line by the base flow profile. Wall temperature very close to the threshold of the transcritical regime are indeed studied here, with $T_w/T_{pb} = 1.04$ being equivalent to the $T_w/T_\infty = 1.15$ case presented in figure 2.

The inviscid growth rate increases as the wall temperature increases in the transcritical regime (figure 3). The cut-off frequency of the instability, at which the perturbation becomes neutral, also increases with T_w, extending the range of unstable frequencies. Note that negative growth rate are found because of the complex mapping mentioned before. Real mappings would only produce neutral modes if no unstable mode exists given the existence of pairs of eigenvalues and their complex conjugate in this case.

It has been seen that the growth rate is small when T_w/T_{pb} is small. In viscous stability calculation, due to viscous damping, this means that this mode could be absent at Reynolds number of the order of those usually studied in boundary layer. In figure 2, a very small portion of the parameter space was unstable at $T_w/T_\infty = 1.15$, while the neutral curve invaded a large region at $T_w/T_\infty = 1.20$. This behaviour is expected for an inflexional instability: the larger the inviscid growth rate is, the more viscous damping is needed to stabilise it, hence the smaller the critical Reynolds number must be. But another viscous effect is actually at play in addition to viscous damping. In figure 4, the growth rate of mode B is plotted for different Reynolds numbers and is compared to the inviscid case. Larger growth rates are obtained for smaller Reynolds numbers presented, indicating a destabilising effect of the viscosity (for

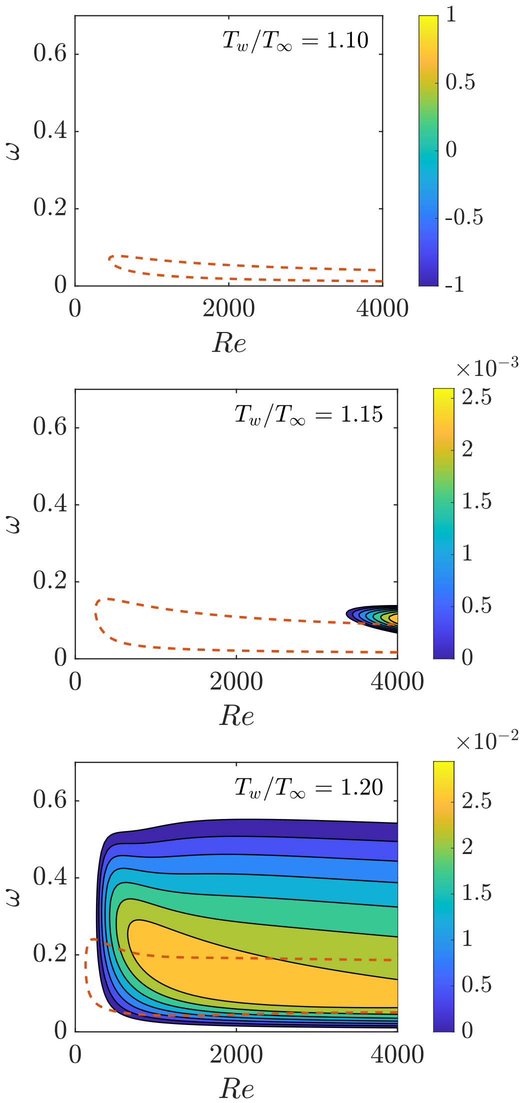

Figure 2: Stability diagram in the Reynolds-frequency space for different wall temperatures. Colours indicate the value of the growth rate of mode B when it is unstable. The red dashed-line shows the neutral curve of mode A, which is unstable in each case. Note that the centre and right plots corresponds to transcritical cases.

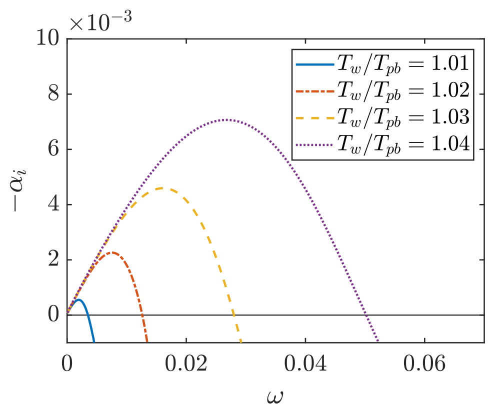

Figure 3: Inviscid linear stability analysis: growth rate for different wall temperatures close to the pseudo-boiling temperature.

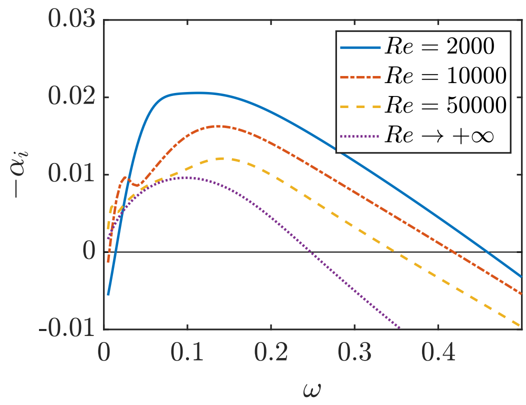

Figure 4: Effect of the Reynolds number on the growth rate of mode B at $T_w/T_\infty = 1.2$. The $Re \to +\infty$ line is obtained from inviscid analysis.

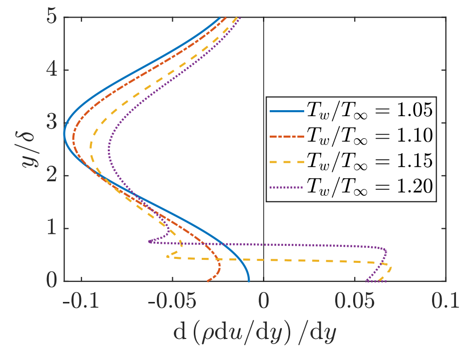

Figure 5: Quantity associated with the generalised inflexion point (GIP) criteria, calculated for different wall temperatures. A GIP is detected when this quantity is equal to zero.

small enough Reynolds numbers, viscous damping eventually takes over and stabilises the flow, as already seen in figure 2). This behaviour will be discussed at the end of the paper as it opens interesting fundamental questions.

ORIGIN OF THE ADDITIONAL UNSTABLE MODE

Discarding viscous effects, the inviscid linearised Navier-Stokes equation can be recast as the Rayleigh equation, solved in the previous section. In the presence of density gradients, it can be shown that a necessary condition of instability is the existence of a GIP in the base flow profile, defined as the location where the quantity $(\bar{\rho}\bar{u}')'$ is zero (the superscript $'$ stands for wall-normal derivative). This quantity is plotted in figure 5. A GIP is found only in the transcritical cases, indicating that some features associated with the Widom line are responsible for it.

To investigate this further, the base flow is examined in the vicinity of the pseudo-boiling point where strong, local gradients of dynamic viscosity are expected. In the boundary layer flow over a flat plate, viscous stresses are usually balanced with advection effects, the latter decreasing and eventually vanishing at the wall. However, if the viscosity is allowed to vary, a term associated with viscosity gradients appears and can, in some region, even be dominant compared to advection. This is more likely to occur near the wall where advection is weak and, of course, for fluids exhibiting large viscosity variations. Under the assumption that viscosity effects dominate the balance of streamwise momentum of the boundary layer equations (the validity of this assumption will be discussed later), we can then write

$$\bar{\mu}'\bar{u}' + \bar{\mu}\bar{u}'' \simeq 0. \tag{1}$$

Furthermore, one can simply rewrite $(\bar{\rho}\bar{u}')'$ as

$$(\bar{\rho}\bar{u}')' = \bar{\rho}\bar{u}'' + \bar{\rho}'\bar{u}'. \tag{2}$$

Combining equations (1) and (2) and making use of $\bar{\mu}' = \bar{T}'(\partial\bar{\mu}/\partial T)$ and $\bar{\rho}' = \bar{T}'(\partial\bar{\rho}/\partial T)$ at constant pressure result in the following approximation:

$$(\bar{\rho}\bar{u}')' \simeq \bar{\rho}\bar{u}'\bar{T}' \left(\frac{1}{\bar{\rho}} \frac{\partial \bar{\rho}}{\partial T}\bigg|_p - \frac{1}{\bar{\mu}} \frac{\partial \bar{\mu}}{\partial T}\bigg|_p \right). \tag{3}$$

Under this approximation, it can be seen that a GIP will appear as soon as the quantity in brackets in the right-hand side is zero. Importantly, this quantity is only related to the fluid properties rather than the flow properties. Noticing that the derivative of the kinematic viscosity ν can be introduced, equation (3) eventually reduces to

$$(\bar{\rho}\bar{u}')' \simeq -\bar{\rho}\bar{u}'\bar{T}' \frac{\partial \bar{\nu}}{\partial T}\bigg|_p \tag{4}$$

This implies that the existence of an extremum of ν is, under the working assumptions, a sufficient condition for a GIP to exist (which is, in turn, a necessary condition for an inviscid instability to exist). The profile of kinematic viscosity is plotted against temperature for real, supercritical fluids in figure 6. All of the presented fluids - O_2, CO_2 and toluene - admit a minimum of ν at the pseudo-boiling line. To verify that this minimum is indeed the key ingredient at the origin of the GIP in supercritical fluids, the approximation obtained in equation (4) is tested in figure 7. It is verified near the wall where viscosity effects are always dominant since advection effects go to zero given the no-slip condition. An excellent agreement is also observed in the immediate vicinity of the pseudo-boiling point, where the above derivation was indeed intended to hold because of the strong gradients of viscosity in supercritical fluids in this thermodynamic region. Crucially, it is at this location that the GIP is found. Therefore, equation (4) indeed provides the interpretation that the GIP originates from the extremum of kinematic viscosity observed at the Widom line. More precisely, it can be shown that a *minimum* of kinematic viscosity is in fact required in order to satisfy the Fjørtoft's theorem extended to varying density fluids. Ultimately, these results suggest that any supercritical fluids, whose flow can be described by the boundary layer approximation and whose temperature profile crosses the Widom line, will exhibit an inviscid instability associated with a GIP located around the Widom line.

DISCUSSION

The additional unstable mode found in the flat-plate boundary layer flow of supercritical fluids has been examined. It has been shown to result from an inviscid instability associated with a GIP that is generated under certain conditions

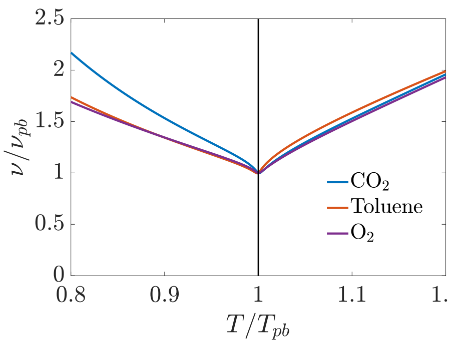

Figure 6: Kinematic viscosity of different supercritical fluids around the pseudo-boiling point ($p/p_c = 1.1$).

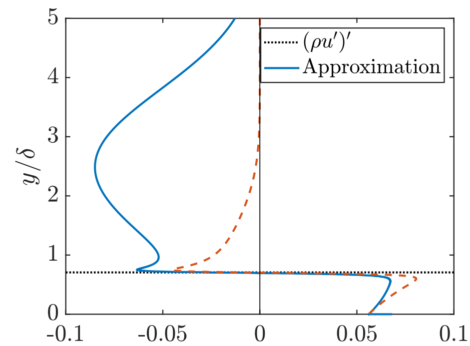

Figure 7: Comparison between the quantity plotted in figure 5 at $T_w/T_\infty = 1.20$ and the approximation proposed in equation (3). The horizontal black dotted-line shows the location of the Widom line.

which supercritical fluids satisfy. These conditions are the presence of strong local gradients of dynamic viscosity coupled with a minimum of kinematic viscosity; no features that are specifically linked to supercritical fluids are eventually invoked. The universality of this result could motivate the investigation of other flows exhibiting these properties. Some fluid mixtures, such as methanol and toluene, are for example known to exhibit a minimum of kinematic viscosity for intermediate mole fractions (McAllister, 1960). The stability of shear flows involving layers of fluids of different viscosities was studied by Yih (1967) who showed that viscosity gradients could cause instability at low Reynolds numbers. Ern *et al.* (2003) investigated this case in the presence of a finite interface between the fluids. This work could be revisited for fluid mixtures featuring a minimum of kinematic viscosity in which an inviscid instability may enrich the dynamics of the system. In this case, it can be noted that the viscosity profile is driven by the concentration of a fluid species, which is a scalar following a transport-diffusion equation analogous to the temperature studied in the present paper. The two cases are therefore very similar from a theoretical point of view.

Different shear flows could also be considered. Plane Couette flow is, for example, expected to lead to similar results as those presented in this work since it strictly satisfies equation (1). This could also constitute a more canonical framework than the flat-plate boundary layer to study mode B. Conversely, plane Poiseuille flow would tell a different story as the streamwise pressure gradient driving the base flow would preclude the use of equation (1) and, subsequently, the validity of the criterion of minimum kinematic viscosity.

Finally, investigating the presence of larger growth at finite Reynolds number than at infinitely large one (observed earlier in figure 4) would be an interesting follow-up study. This phenomenon is not unusual as it occurs, for example, with the first mode of a supersonic boundary layer which, in addition of being inviscid unstable, bears the viscous destabilisation mechanism of the incompressible TS waves (see for example Mack (1984)). The TS-viscous mechanism is already associated with mode A in supercritical fluids. This opens fundamental questions regarding the destabilisation mechanism of mode B at finite Reynolds number, which would merit a dedicated investigation. Given the aforementioned similarity with miscible fluids, a potential approach could be to study if the low Reynolds number instability existing in miscible fluids could, to some extent that should be understood, be invoked to explain this behaviour.

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