A MOLECULAR BASIS FOR THE
ONSET OF TURBULENCE

AMADOR C. MURIEL

Center for Theoretical and Computational Sciences
(World Laboratory), SBMA, Olangapo, Philippines

ABSTRACT

We analyze existing data on the onset of turbulence for pipe flow in the light of a molecular theory for the onset of turbulence. In contrast to the use of derived quantities for characterizing the onset of turbulence, such as the Reynolds number, we restrict our data and analysis using observable quantities such as molecular properties and critical velocities. We find a rationale for the molecular origin of turbulence, in contrast to the exclusive use of continuum theory.

There now exist some recent data for studying the onset of turbulence in pipe flows. In the first experiment (Nerushev & Novopashin, 1996, 1997), it was asserted that there are two distinct critical Reynolds numbers for the onset of turbulence in a Pouseuille-Hagen pipe flow experiment in pure carbon monoxide and nitrogen. Since the fluid properties of these two gases are alike, this result was viewed with some suspicion. In a second experiment that attempted to duplicate the first, it was asserted that there was no meaningful difference between the critical Reynolds numbers for argon, carbon monoxide, and nitrogen (White & Sreenivasan, 1998). In a third experiment, different critical Reynolds numbers were detected for the noble gases helium, neon, argon, krypton, and xenon but was since repeated under changed circumstances that reduced the differences of the critical Reynolds numbers. The above results are not considered definitive because of the compressible nature of efflux from a pressurized vessel. Indeed, when the results are displayed traditionally in the form of Reynolds numbers, they are subject to uncertainty due to density and viscosity corrections. To reduce the influence of compressibility in what is certainly a non-equilibrium process, a fifth experiment has been done on liquids: normal water, heavy water and methyl alcohol (Novopashin, to be published), opening the question of the onset of turbulence for incompressible and isothermal liquids.

The Reynolds number is defined as $\rho v d / \eta$ where ρ is the density, v is the mean velocity, d is a typical, but somewhat arbitrary apparatus dimension, and · is the dynamic viscosity. This dimensionless parameter is taken as an index for the laminar-turbulent flow transition. It derived its significance from a redimensioned Navier-Stokes equation, leading to a general principle called Reynolds invariance or scaling (Frisch, 1995). The long accepted tenet is that the critical Reynolds number should be independent of specific molecular constituents; for as long as both the density and viscosity is specified, there is no longer a need to identify the molecules. In this light, the critical Reynolds number should be the same whether air, nitrogen, carbon dioxide, or water, is used in an experiment, a strong form of invariance in continuum dynamics based on the Navier-Stokes equation. An assertion to the contrary has to be experimentally tested, hence the impetus to do all five experiments quoted in this paper.

The theoretical basis for asserting that the onset of turbulence is molecule dependent is simple. To begin with, continuum hydrodynamics is based on the aggregation of point particles in a fluid into a continuous medium. Both energy and momentum are conserved in molecular collisions, so the contribution of molecular collisions to fluid flow are not considered important. But this is not so when molecules collide inelastically. In laminar flow, the average energy of collisions is too low to render molecular collisions inelastic. But one can calculate the average kinetic energy of molecules in fluid flow to see that at some energy density in a fluid, the internal degrees of freedom of molecules may be affected. This means that energy may be absorbed or radiated in a strongly agitated system. To illustrate:

(1)   Diatomic molecules in their ground states have rotational energies of the order of 8×10^{-16} erg (5.03×10^{-4}ev). Note also that room temperature kT is roughly 4.09×10^{-14} erg (2.57×10^{-2}ev). We can see that rotational energy excitations and de-excitation are well within the range of a room temperature gas. We will expand on this point later.

(2)   An argon atom participating in fluid flow with an average velocity of 20cm/sec has a kinetic energy of 1.33×10^{-20}erg (8.4×10^{-9}ev). Increasing the velocity to 350cm/s will increase the energy to 10^{-18}erg (10^{-6} ev) -- this energy is well above the energies of quantum confinement states in the cell model of a gas. As is well-known, quantum confinement arguments in gases, where structureless atom is boxed by a neighboring gases, can completely recover classical thermodynamic results for the ideal gas (Hill, 1960).

When mechanical or thermal agitation of a fluid is sufficiently strong, the internal degrees of freedom of molecules are excited. Part of the kinetic energy of motion of molecules arising from fluid flow is converted to energies of collision. If this energy is sufficient, a diatomic molecule, for example, could become even

more excited, and give up radiation in the process - a turbulent fluid is known to heat up and dissipate energy (Rivera & Li, 2000).

To explain our rationale, consider a diatomic molecule with rotational states. An excited diatomic molecule gives up radiation, ultimately a quantum phenomenon with built in stochasticity, the direction of the emitted radiation can only be described probabilistically. By conservation of momentum and energy, the molecules recoil stochastically as well. The stochasticity we are discussing here is different from the stochasticity contributed by an external heat bath. We identify this small stochastic contribution to molecular recoil velocities as the seed for the onset of turbulence. This stochastic variety will be propagated and magnified in a turbulent fluid. In this way, we find a physical origin for the Reynolds decomposition of fluid velocity into a regular and stochastic component (McCumb, 1961). Heretofore, the Reynolds decomposition has been justified by the existence of turbulence, one must acknowledge its stochastic contribution by inserting the decomposition in the Navier-Stokes equation. It is also a mathematical approach that does not prescribe from first principles when such a decomposition begins. In contrast, we propose here that the Reynolds decomposition be dictated by the physics of excitation. Historically and mathematically, it is only after the Reynolds decomposition is assumed that one can begin to model turbulence. For this reason, we suggest that the Navier-Stokes equation, alone, cannot describe the entire range of laminar, laminar-turbulent and turbulent regimes, without inserting turbulent-like solutions first. In fact, this is how turbulence is characterized in numerical simulations (Deissler, 1984).

Of course there is the case of noble gases which are simple enough to be modeled as point masses participating in fluid flow, they too become turbulent without the use of an internal degree of freedom such as may be found for diatomic molecules. We shall address this problem first. To look into the case of noble gases, we look back at a very early and successful application of quantum mechanics to the ideal gas, the cellular model (Hill, 1960). This model assumes that a molecule or atom is caged by a box of other molecules, which is a simple cube of dimension $L = (m/\rho)^{1/3}$ where m is the molecular mass and ρ is the density. This density is the density of a quiescent gas, not a turbulent gas, this point is important. From the quantum mechanics of boxed particles, the energy of this molecule is given by

$$E = \frac{h^2(n^2 + m^2 + l^2)}{8mL^2}$$

where the quantum numbers n, m, l run from 1 to infinity and h is Planck's constant. Using the quantum states above, the thermodynamics of an ideal gas has been completely derived from textbooks.

We now use the cell model as a mental construct to explain the onset of turbulence. Consider an atom caged by its surrounding molecules. The cage and molecule inside may be moving bodily with a fluid velocity, n. When the fluid velocity is increased, part of the molecular velocities is converted to kinetic energies of collision. According to our picture, part of this kinetic energy of motion is converted to mutual collisions which result in the excitation of the caged molecule as a result. Although such collisions are obviously complex, we continue to adopt the cell model but introduce the excitation of the caged molecule. This allows the caged molecule to be excited to an internal energy state given by E. Using the box dimension above, the lowest energy transition of the caged molecule from the ground state is given by:

$$\Delta E = \frac{3h^2}{8m} \left(\frac{r}{m} \right)^{2/3} \qquad \qquad 1.$$

which will have to be overcome by the energy due to collisions. According to our model, a critical fluid velocity is needed to overcome this energy, given by

$$\alpha m v^2_c = \Delta E \qquad \qquad 2.$$

where α expresses the unknown fraction of the energy drawn from the fluid kinetic energy that is converted to the excitation energy of the molecule. The fraction α is dependent on the geometry of molecules as well as the boundary condition of the experiment. This equation connects the macroscopic fluid velocity to the internal energy states of the molecules. In classical fluid flow, $\pm$ is zero.

The excitation energy in Eq. (2) is later given up as radiation. We can then use

$$\Delta E = hc/\lambda \qquad \qquad 3.$$

where h is Planck's constant. c is the speed of light, and λ is the wavelength of the ensuing radiation.

For noble gases, the internal energy is provided by quantum confinement as in the cell model. Substituting the gap energy, w get the following critical velocity:

$$4.$$

$$V_c = \frac{h}{m} \left(\frac{r}{m} \right)^{1/3} \sqrt{\frac{3}{8\alpha}}$$

Parenthetically, from Eq. (4) we may obtain a non-unique critical Reynolds number

$$Re_c = \frac{\rho v_c d}{\eta} = \frac{hd}{\eta}\left(\frac{\rho}{m}\right)^{4/3}\sqrt{\frac{3}{8\alpha}} \qquad\qquad 5.$$

where d is a typical dimension of the turbulence apparatus, rather arbitrary, as remarked earlier, and h is the dynamic viscosity. The critical Reynolds number is not a fundamental dimensionless constant such as the fine structure constant of quantum mechanics.

If we now perform identical laminar-turbulent transitions experiments on two different pure noble gases using the same apparatus, we can form a ratio

$$\frac{Re_{c1}}{Re_{c2}} = \left(\frac{\rho_1}{\rho_2}\right)^{4/3}\left(\frac{m_2}{m_1}\right)^{4/3}\left(\frac{\eta_2}{\eta_1}\right)\left(\frac{\alpha_1}{\alpha_2}\right) \qquad\qquad 6.$$

Now if the molecular geometry is the same, the two a's cancel, and we get the following relation for the noble gases.

$$Re_c h(m/r) = Re_c v m^{4/3}/r^{1/3} = constant \qquad\qquad 7.$$

where v is now the kinematic viscosity. Were the data reduction simple and straightforward, Eqs. (6,7) are immediately testable. In practice, one has to correct the density and viscosity by temperature changes, but in a process where the flow is neither isothermal nor imcompressible, such corrections are not really straightforward. We point this out because turbulence is a non-equilibrium thermodynamic concepts of temperature and pressure are not rigorously defined. It is best to test Eq. (4) in the form

$$\frac{v_{c1}}{v_{c2}} = \left(\frac{\rho_1}{\rho_2}\right)^{1/3}\left(\frac{m_2}{m_1}\right)^{4/3} \qquad\qquad 8.$$

for two monoatomic gases with identical geometry and apparatus. There are no adjustable parameters in this estimated ratio, the densities are obtained from quiescent gases because the densities enter by way of the dimension of the hypothetical cell. At the onset of turbulence, there need not be corrections due variable densities (and viscosities) in a subsequent non-equilibrium efflux from a pipe. The densities used in Eq. (8) are static densities of the gases measured in our cell model in thermodynamic equilibrium, whereas the densities in Eq. (7) are supposed to be the instantaneous densities of the flowing gases. Therefore, we have taken the data from all experiments from Novosibirsk and Lyon to get the raw measurements of the critical velocities for the noble gases and obtain the following ratios in Table I:

Ratio of critical velocities	Experiment	Theory Eq. (8)
$\dfrac{v_{He}}{v_{Ne}}$	3.03 [4]	5.04
$\dfrac{v_{He}}{v_{Ar}}$	9.03 [4]	9.97
$\dfrac{v_{He}}{v_{Kr}}$	19.8 [4]	20.94
$\dfrac{v_{He}}{v_{Xe}}$	33.72 [4]	32.72
$\dfrac{v_{He}}{v_{Ne}}$	0.33 [4] 0.34 [6]	0.336

Ratio of critical velocities	Experiment	Theory Eq. (9)
$\dfrac{v_{N2}}{v_{CO}}$	0.92 [1]	0.88
$\dfrac{v_{CO2}}{v_{N2}}$	0.517 [6]	
$\dfrac{v_{SF6}}{v_{CO2}}$	0.290 [6]	
$\dfrac{v_{N2}}{v_{O2}}$	1.02 [6]	0.99

Ratio of critical velocities	Experiment	Theory Eq. (9)
$\dfrac{v_{D2O}v_{H2O}}{v_{CH3O3}v_{H2O}}$	1.18 [5]	1.10

For more complex gases, we obtain the data from Novosibirsk (Nerushev and Novopashin, 1996, 1997) Yale (White & Sreenivasan, 1998), and Lyon (Sommeria, unpublished).

Now for the unresolved Novosibirsk (Nerushev and Novopashin, 1996, 1997) and Yale (White & Sreenivasan, 1998), experiments. According to the above cell model interpretation, the critical velocities for carbon monoxide and nitrogen should be the same. They have the same molecular mass, density, as well as dynamic viscosity. Both the critical velocities and critical Reynolds number should be the same, from classical continuum dynamics, and even from the above quantum confinement arguments. However, the rotational energies of these diatoms are lower than the confinement energies, they are more accessible and get excited first. So it is the rotational energy which must be considered. Now carbon monoxide and nitrogen have different rotational energy spectra (they have different parity), so the threshold for turbulence will be different. According to our picture, it is the

gas whose molecules possess a lower excitation state which will get turbulent sooner, in this case, nitrogen. Hence the quantum confinement argument for noble gases is replaced by rotational excitation to display a difference in the onset of turbulence. In fact, diatomic molecules at room temperature already possess occupied excited states. So our early example of the excitation of the ground state due to collisions will have to be modified by fairly complex calculations. Fortunately the resulting radiative transitions in an agitated real gas is already available without the need to calculate them. If one looks at the infrared spectra of nitrogen and carbon monoxide, we find that wavelengths corresponding to the lowest and most excitable internal energies are respectively $\lambda_{N2} = 8722$ Angstroms and $\lambda_{CO} = 6804$ Angstroms, respectively (Lide, 1999). These dominant transition wavelengths would be ideally the result of a complex many-body calculation, fortunately available to us as spectroscopic observations.

From Eq. (2) and (3), we form the ratio

$$\frac{v_{c1}}{v_{c2}} = \left(\frac{\lambda_2}{\lambda_1}\right)^{1/2}\left(\frac{m_2}{m_1}\right)^{1/2} \qquad\qquad 9.$$

where λ is the wavelength of the radiation from the excited state obtainable from spectral tables. This formula is useful for complex molecules. Again, there are no adjustable parameters. The experimental and theoretical results for carbon monoxide and nitrogen are also included in Table 2.

It appears that the rationale are as follows. In any gas or liquid, we must look for the lowest form of energy excitations, this may well be quantum confinement of simple atoms, or molecular rotation. Once the lowest energy spectrum is determined, we use that to define the onset of turbulence. But whatever it is, the onset of turbulence seems to have a molecular, and therefore quantum origin.

Let us now move on to more complex molecules, in liquids, such as normal water, heavy water, and methanol. As it turns out, the quantum confinement argument discussed above introduces a gap energy that is higher than the internal deformation, or vibrational states of molecules. By our prescription of utilizing the smallest energy gaps, we find that quantum confinement energies are too large, while the vibrational states corresponding to bending or deformation of the molecules require less energy. As we remarked earlier, these lowest identifiable energies are what determines the onset of turbulence.

For normal water, the wave number of radiations is $1/\lambda = 159800$ waves per meter, while for heavy water it is 117800 waves per meter. The ratio of critical velocities for heavy and normal water is calculated from Eq. (9) using the deformation states of water, to give 1.10 versus the experimental value of 1.18. We display the results for liquids in Table 3.

For many complex liquids, where quantum confinement is not relevant for the laminar-turbulent transition, the best tool in calculating ratios of critical velocities is the infrared spectrum available in handbooks (Sigma-Aldrich Co., 1997).

All of the above arguments have originated from rather involved theoretical considerations (Muriel and Dresden, 1995-1997; Muriel *et al.*, 1998; Muriel, 1998), which provided the impetus to perform the experiments. Once this phase of theorizing and experimentation has begun, as in fact it has, it is possible to explore new features of this view if the onset of turbulence, even independently of our the original theoretical work. It is difficult to accept that the data above are merely coincidental. We can thus draw a weak statement of our thesis that molecular properties affect the onset of turbulence. When properly scaled by the pipe diameter, density and viscosity, the above data translate to different critical Reynolds numbers for different fluids. The strong statement, which must be further tested, is that quantum structure determines the onset of turbulence.

We offer in this Letter some ideas on how the onset of turbulence maybe further studied:

1. The Yale experiment studied two like gases – carbon monoxide and nitrogen, and a noble gas, argon. In the light of the Novosibirsk results on noble gases, it might be useful to repeat the Yale experiment on more noble gases, instead of just argon. A difference in the critical Reynolds number of 6% for carbon monoxide and nitrogen was in fact seen, and adjudged insignificant because of experimental errors.

2. Of all the experiments performed to date, only the Lyon tests by Sommeria directly measured velocity, while other experiments inferred the velocities from continuity arguments and the ideal gas law. Most of the gases are not ideal and the second virial coefficients have some significance as observed by Novopashin & Muriel (1998)

3. The classic quasi-static approach of Novopashin and the dynamic free efflux experiment of Sommeria (unpublished) using ultrasound techniques may now be augmented by other experiments, like the measurement of the radiation spectrum of a turbulent gas, and the role of radiation on the onset of turbulence, two other effects that we have predicted earlier (Muriel and Dresden, 1995-1997; Muriel *et al.*, 1998; Muriel, 1998).

When these are done, it is possible to introduce to turbulence research some new techniques from ultrasound and molecular spectroscopy which can only enrich the field, and cultivate the hope of explaining the turbulence from first principles of physics, instead of from the mathematics flow of phenomena alone.

ACKNOWLEDGEMENT

REFERENCES

Deissler, R.G. 1984. *Review of Modern Physics* 56 223.

Frisch, U., 1995. Turbulence: The legacy of A.N. Kolmogorov, Cambridge University Press, Cambridge.

Hill, T.L. 1960. An introduction to statistical thermodynamics, Dover Publications, New Your.

Lide, D. (Ed), CRC Handbook of Chemistry and Physics, 80th Edition, CRC Press.

McComb, W.D., The Physics of fluid turbulence, Clarendo Press, Oxford, 1961.

Muriel, A. 1998. *Physica D*, 124:225.

Muriel, A. and M. Dresden. 1995. *Physica D*, **81**: 221.

Muriel, A. and M. Dresden. 1996. *Physica D*, **94**: 103.

Muriel, A. and M. Dresden. 1997. *Physica D*, 101, 299.

Muriel, A., P. Esguerra, L. Jirkovsky, M. Dresden. 1998. *Physica D*, 119, 381.

Nerushev, O.A. and S.A. Novopashin. 1996. *JETP Letters*, 64: 47-50.

Nerushev, O.A. and S.A. Novopashin. 1997. *Physics Letters*, *A*, 232-243.

Novopashin, S.A. and A. Murie. 1998. *JETP Letters*, **68**, No. 7, 582.

Novopashin, S.A. and A. Muriel. 2000. *Technical Physics Letters*, 26(3): 231-232.

Rivera, M., X.Li. 2000. Revise Letters 85, 976.

Sigma-Aldrich Co. 1997. The Aldrich Library of FT-IR Spectra, V. 1.

White, C.M. and K.R. Sreenivasan. 1998. *Physics Letters A*, 238: 323-327.