

A Century of Transport – A Personal Tour

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The industrial chemists and mechanical engineers who banded together in 1908 to form the AIChE were self-taught experts in the design and operation of equipment to carry out chemical processing. Although graduates with a degree in chemical engineering had been entering industry for over a decade, none of them were among the founding members because they failed to meet the qualification of “years of responsible charge”. The skills possessed by the founding members can be discerned from the early identification of chemical engineering with what we now call the *unit operations*. Remarkably, these unit operations were all physical and did not invoke chemical reactions explicitly.

The work of the first chemical engineers could be distinguished from that of chemists, other than perhaps “industrial” ones, by their utilization of continuous processing and thereby of an Eulerian rather than a Lagrangian framework for modeling, as well as by their involvement with transport, in particular with fluid flow, heat transfer, and mass transfer. Their work could be distinguished from that of mechanical engineers by virtue of their involvement with chemical conversions and chemical separations, and over the century with an increasing variety of materials, many of them synthetic and new to the world. Accordingly, the differences in the approach of chemical engineers and mechanical engineers to fluid mechanics, heat transfer, and thermodynamics have continued to widen.

The joint designation of fluid mechanics, heat transfer, and mass transfer as *transport* rather than as *unit operations* and/or as separate topics, began with the publication in 1960 of the most influential book in the history of chemical engineering, namely *Transport Phenomena* by Bob Bird, Warren Stewart, and Ed Lightfoot. Most advances come at a price, and in the long run this consolidation and theoretical upgrading has resulted in a lesser preparation of our students in applied fluid mechanics and heat transfer *per se*.

Although our understanding of transport has greatly progressed over the century, this subject now has lesser role in education and practice, because the total number of technical courses in the engineering curriculum has regularly been decreased, and because additions of new subject matter have led to a reduction in those devoted to engineering science. These “advances” have come at the price of a poorer preparation for our students in transport itself. The current “in” topics of biotechnology and nanotechnology involve transport but mostly at such a smaller scale that what I will be describing is applicable, if at all, only as a guideline.

My objective today is to review the evolution of the skills and practices of chemical engineers in dealing with transport while avoiding, insofar as possible, those aspects being addressed by the other speakers, namely the validity of the Navier-Stokes equations, the role of numerical computation, and biological applications.

Because of the aforementioned expansion of knowledge and applications in transport, its breadth, even in a classical sense, is such that I cannot cover all aspects or even the evolution of any one aspect completely. My coverage today is therefore both highly selective and very arbitrary. My initial plan was to trace developments in 1) turbulent flow and convection in tubular flow, because much chemical processing occurs in that regime and geometry, 2) transport in packed and fluidized beds, because the advances have been made almost exclusively by chemical engineers, 3) mass transfer, because chemical engineers

have an almost exclusive role therein, 4) non-Newtonian flow, because chemical engineers have a large although not exclusive role therein, and 5) combustion, because chemical engineers appear to have the most appropriate background.

However, this seemingly logical approach proved to be awkward to implement, and after a struggle with it, I switched to the evolution and adaptation of critical concepts and methodologies, namely the identification of compound variables, irrespective of the process itself, and the development of predictive expressions, correlating equations, and analogies. The path of progress itself is traced for only two topics in transport, namely turbulent flow and turbulent convection, and even then only in terms of a few highlights. My own primary field of research, namely combustion, fell by the wayside. I conclude with some general observations concerning the influence of AIChE programming at its meetings, and of the sources and extent of funding for advances in transport.

The date of 1908 is a signal one for us here today, but it is not a dividing line in terms of chemical technology. Many of the concepts that now characterize the contributions of chemical engineers to society originated earlier. Also, as painful as it is to admit, many of our treasured and most useful concepts originated, even after 1908, with non-chemical engineers. Rather than be chauvinistic, I have chosen to identify the actual origins of these concepts and to relate our subsequent applications and improvements to those forerunners and professional associates, even though they did not have the wisdom or foresight to identify themselves as chemical engineers.

My choice of and order of individual topics is not important because they are intended merely as illustrations. I concede and apologize that several of the choices were influenced by my own involvement – not because of that involvement but because I am more confident in and ready access to the historical record in those instances. In defense of that position, I may cite my presence on the sidelines or in the arena of chemical engineering for 88 of the last hundred years, my commitment to chemical engineering as a profession for 70 of them, my formal association with the AIChE for 69, and my focus on transport over most of that span.. Of course these experiences do not qualify me to predict the future direction of transport, so I will stick to my assigned topic – a review and interpretation of progress during *The First Century of the AIChE* and leave the predictions to others. .

I am taking one liberty that may run contrary to the mood of this celebration of our roots in transport, namely citations of wrong turnings. Such identifications are not meant to impugn those who originated the false or inferior concepts and expressions, because all concepts in science and engineering are eventually discarded or improved upon; that is the process by which progress is made. Indeed, the original concepts and expressions that are later proven wrong are in most instances a necessary first step, and their discard usually reflects the acquisition of new information or a new insight.

I have taken one other liberty, namely favoring technical continuity over strict chronology. The resulting order of topics might be confusing to undergraduate students but not to this audience because the topics taken out of temporal sequence are familiar ones.

Continuity and Conservation

The equations of conservation – the Navier-Stokes equations and their counterpoints for energy and species – are the starting point for most theoretical work on transport, but, out of deference to Howard Brenner, I will not trace the development of these equations nor examine their validity except to mention one contrary opinion that brought me up short at the time. The renowned physicist George E. Uhlenbeck, whom I was fortunate enough to have as a mentor, was so frustrated by his failure to confirm or disprove the N-S equations by reference to statistical mechanics, which he considered to be a better starting point, once

wrote “Quantitatively, some of the predictions from these equations surely deviate from experiment, but the *very* remarkable fact remains *that qualitatively* the Navier-Stokes equations *always* describe physical phenomena sensibly.....The mathematical reason for this virtue of the Navier-Stokes equations is completely mysterious to me.”

Conceptual and Compound Variables

The use of conceptual and compound variables in chemical engineering is so pervasive that we may forget that they are arbitrary, that in many instances their introduction was a consequence of great ingenuity, and that we almost blindly place our trust in the unexplored variables buried within the compound ones. I will call your attention to a few of them.

The heat transfer coefficient The heat transfer coefficient is perhaps the best example of a revolutionary concept and of a compound variable that has become imbedded in chemical engineering. Isaac Newton in 1701 noted that the rate of heat transfer to a surface in free convection was proportional to the area times the difference between the free-stream temperature and the temperature of the surface. He thereby inspired the replacement of four variables with one, namely the coefficient of proportionality that we now call the *heat transfer coefficient*. This concept, although usually encompassing a degree of approximation and scorned by some, has remained in active use for over 300 years. Eventually it was extended to forced convection in tubular flow by replacing the free-stream temperature with the mixed-mean temperature of the fluid, and was adapted for mass transfer in terms of fugacities, partial pressures, and/or concentrations. The friction factor, drag coefficient, and orifice coefficient are closely related conceptual variables for momentum transfer. These five coefficients have been utilized by chemical engineers for more than one hundred years, and they remain invaluable and irreplaceable. Most of our technical data-base is compiled in this form.

The equivalent thickness for pure conduction This quantity, as defined by $\delta_e = j_w/k\Delta T$ (or h/k) has repeatedly been proposed as an alternative to the heat transfer coefficient, but, with one notable exception, it has always proven inferior in terms of correlation, generalization, and insight. That exception was its use by Langmuir in 1912, in the context of an analysis of heat losses by free convection from the filament of a partially evacuated electrical light bulb. He utilized the concept of an equivalent thickness for thermal conduction to derive an approximate expression for the effect of the curvature of the surface of the wire, *vis-à-vis* a vertical flat plate, on the rate of heat transfer by free convection. The resulting expression, which, after nearly a century, has not been improved upon and which is based on the applicability of the log-mean area for conduction across a cylindrical layer, is

$$Nu = 2 / \ln \{ 1 + (2 / Nu_f) \}$$

Here Nu_f is a correlating equation for free convection from a vertical flat plate. It should be noted that the effective thickness has vanished from the final expression. This relationship has been found to be uniquely useful as an approximation for both free and forced convection from a horizontal cylinder as the Grashof and Reynolds numbers, respectively, approach zero, and is readily adapted for the region of the entrance in laminar tubular flow and for mass transfer. When applied to a spherical layer this concept invokes the geometric-mean area and leads to an exact asymptote for a decreasing Reynolds number or Grashof number. “*Vive la exception*” due to Langmuir.

The mixed-mean velocity The mixed-mean velocity has proven to be a very useful concept in chemical engineering because of the pervasive use of tubular flow in chemical processing. For constant density it is defined as

$$u_m \equiv 2 \int_0^1 u_r \left(\frac{r}{a} \right) d \left(\frac{r}{a} \right)$$

For fully developed flow, the mixed-mean velocity is simply equal to v/A_x , where v is the volumetric rate of flow and A_x is the cross-sectional area. The integration is thereby rendered unnecessary in so far as v is known.

The mixed-mean temperature The mixed-mean temperature has also proven to be a very useful quantity in chemical engineering for the same reason. For a fluid of constant density and heat capacity in a round tube it is defined as

$$T_m \equiv 2 \int_0^1 T_r \left(\frac{u_r}{u_m} \right) \left(\frac{r}{a} \right) d \left(\frac{r}{a} \right)$$

There is, however, no directly and simply measurable quantity for the mixed-mean temperature analogous to v/A_x for the mixed-mean velocity. I will later describe one instance in which this concept has led us astray for nearly a century, but that exception does not undermine its overall usefulness.

Mixed-means in general The corresponding expression for the mixed-mean concentration can readily be inferred, and the slightly more complex expressions for the mixed-mean velocity, temperature, and concentration in the cases of variable density, heat capacity, and/or concentration can readily be formulated, but they are less useful because of their relative complexity and the difficulty posed by their measurement.

Fully developed flow The velocity profile develops down-stream from the entrance to a tube and eventually approaches an asymptotic one in both the laminar and turbulent regimes. The local value of the friction factor decreases and approaches an asymptotic value in laminar flow and in turbulent flow in a rough but not a smooth pipe. This asymptotic state for the velocity distribution is classified as *fully developed flow*. The rate of development depends on the geometrical configuration and the Reynolds number, and in the turbulent regime the asymptotic velocity distribution itself also depends on the Reynolds number. In tubes with a large ratio of length to diameter, the concept of fully developed flow is often utilized as an approximation for the entire length of the tube. Strictly speaking, fully developed flow implies constant density and viscosity.

The friction factor for artificially roughened tubes In 1932 Nikuradse, a student of Prandtl, confirmed experimentally beyond all question that the friction factor for turbulent flow in a smooth pipe decreases indefinitely as the Reynolds number increases. In 1933 he discovered that if an artificial uniform roughness is imposed on the surface of the pipe the friction factor instead approaches an asymptotic value. He devised a correlating equation for this asymptotic value as a function of the amplitude of the roughness divided by the diameter. Although the results of this experiment and analysis were of no direct practical value because the roughness in commercial piping is anything but uniform, this work of Nikuradse not only provided insight but was an essential precursor for the practical developments that followed.

The friction factor for commercial (naturally roughened) tubes Colebrook in 1938-39 reported on his measurements of the friction factor for real piping of various types, and proposed a truly ingenious concept for the representation of their "effective" roughness, namely the value that produced the same asymptotic friction factor as that given by the aforementioned correlating equation of Nikuradse for uniform roughness. The practical utilization of this concept requires a tabulation of effective values of the roughness for various types of piping, such as steel, both new and aged and glass. (As an aside, the tables of roughness in most of our current textbooks and handbooks are badly out of date and should

be revised to reflect modern processes for the manufacture of tubing.) Colebrook also formulated a generalized empirical equation for the approach to that asymptotic value as the Reynolds number increases. The turbulent regime of every friction factor plot that you have ever utilized is merely a graphical representation of that equation.

The equivalent length A similar concept to that of the effective roughness has long been utilized to estimate the pressure drop due to pipe fittings such as valves and elbows, namely the length of straight smooth pipe that results in approximately the same pressure drop for a given rate of flow. This concept implies that the dependence of the pressure drop on the rate of flow is the same for the fitting as for straight piping, which is a crude approximation at best. The application of this concept for design requires a table of values. Again, the tables in our handbooks are out-of-date and of unknown reliability.

“Plug flow” An unfortunate concept pervades much of the literature of chemical engineering, and in particular that of reactor engineering, namely “plug flow”, which occurs physically only when some semi-soft solid such as ice cream is pushed through a tube by a plunger. Turbulent flow does not approach “plug flow” as the Reynolds number increases; the velocity still goes to zero at the wall and the velocity profile is better characterized as parabolic-like than as flat. The idealization of “plug flow” does result in simple and reasonably accurate solutions for the chemical conversion for some conditions, but also very erroneous ones for others. The approximate solutions for chemical conversions that are based on “plug flow” need not be discarded; they can simply be reinterpreted as those for a physically conceivable condition, namely perfect radial mixing, that is for $Pr \rightarrow 0$ and $Sc \rightarrow 0$. Solutions for “plug flow” appear in the literature of heat transfer, but only in the context of lower bounds for Nu .

Integral boundary-layer theory When I was a graduate student, the advanced textbooks on fluid mechanics and heat transfer included a section on integral-boundary-layer theory. This methodology, which was apparently devised independently by von Kármán and E. Pohlhausen in 1921, was based, for flow, on the postulate of an arbitrary velocity distribution, thus allowing an analytical integration of the momentum balance to obtain a closed-form expression for the drag coefficient. The equivalent process was applied for the temperature distribution to obtain an expression for the heat transfer coefficient. One positive measure of progress in the field of transport is the disappearance of this concept from modern books on fluid mechanics and heat transfer.

Potential (inviscid) flow and the boundary layer concept Boussinesq in 1905, and thus just three years before the founding of the AIChE, derived solutions for forced convection in inviscid flow over immersed bodies. These solutions are now recognized to have practical value only as asymptotes for $Pr \rightarrow 0$ and in the boundary layer model of Prandtl, in which the latter conceived of a thin layer of slowly moving fluid next to the surface and inside a regime of inviscid flow. This concept remains alive today, but its limitations should not be overlooked.

Free streamlines The boundary between moving and non-moving segments of a fluid stream is called a *free-streamline*. The pressure is constant along such a line or surface. This concept is an important one in aerospace engineering and in civil engineering, and has at least one lasting application in chemical engineering, namely the prediction of a value of 0.5 for the coefficient for both a planar and circular Borda entrance, and a value of $\pi/(2+\pi) = 0.6110\dots$ for both a planar and a circular orifice. These values are, however, only approximations. For example, the actual limiting value for a sharp-edged orifice in a round tube is 0.5793 rather than 0.6110.

Criteria for turbulent flow In 1883 Osborne Reynolds, on the basis of the most famous experiment in the history of fluid mechanics, deduced that below a certain numerical value of the dimensionless group Du_{mp}/μ , now named for him, a perturbation introduced into a fluid

flowing through a pipe would dampen out. That value is generally taken to be 2100. However, the value below which the flow is totally free of turbulent fluctuations, namely $Re \cong 1600$ or $a^+ = a(\tau_w \rho)^{1/2} / \mu = Re(f/8)^{1/2} \cong 56.6$, and that above which the flow is fully turbulent, namely $Re \cong 4020$ or $a^+ = Re(f/8)^{1/2} \cong 150$, are of more practical utility in terms of transport.

Fully developed convection The concept of fully developed convection is much more subtle and its initial formulation required some ingenuity. The temperature of the fluid stream in a heated or tube increases continuously but, by virtue of the nearly linear dependence of the enthalpy on temperature, a dimensionless temperature that approaches an asymptotic value may be defined for some particular thermal boundary conditions.

For example, if a uniform heat flux density is imposed on the fluid at the wall, the mixed-mean temperature of the fluid must increase linearly with axial distance insofar as the density and heat capacity can be considered invariant. It follows that the temperature of the wall must thereafter also increase linearly at the same rate, and that $(T-T_0)/(T_m-T_0)$, $(T_w-T)/(T_w-T_m)$, and the heat transfer coefficient must approach asymptotic values. Fully developed convection for uniform heating is thus defined by the near-attainment of asymptotic values for $(T-T_0)/(T_m-T_0)$, $(T_w-T)/(T_w-T_m)$ and the heat transfer coefficient. It follows that $\partial T/\partial x \rightarrow \partial T_w/\partial x \rightarrow \partial T_m/\partial x \equiv dT_m/dx$, which allows simplification of the differential energy balance.

A uniform wall-temperature also results in an approach to an asymptotic value for the heat transfer coefficient and for $(T_w-T)/(T_w-T_m)$ but not for $(T-T_0)/(T_m-T_0)$. Seban and Shimazaki in 1951 recognized that the attainment of an asymptotic value of the first of these quantities implied that its derivative could be equated to zero. They thereby derived

$$\frac{\partial T}{\partial x} = \frac{\partial T_m}{\partial x} \left(\frac{T_w - T}{T_w - T_m} \right)$$

The two terms on the right-hand side are both independent of the radius, and therefore $\partial T/\partial x$ can be replaced by dT/dx . Substitution of this expression in the differential energy balance results in some simplification.

Fully developed convection is often utilized as an approximation for the entire length of the tube. Most analytical and numerical solutions for thermal convection are for fully developed convection and thereby for uniform heating or uniform wall-temperature because of the relative simplicity of the behavior. The former condition can be approximated in practice by electrical-resistance heating of the tube wall or by equal counter-enthalpic flow, and the latter condition by subjecting the outer surface of the tube to a boiling fluid (or for cooling to a condensing fluid). Constant viscosity, as well as density and heat capacity, are implied by the concept of fully developed thermal convection. The concept of fully developed mass transfer can readily be formulated using fully developed heat transfer as a guide.

The Boussinesq transformation The combination of approximations and changes of variable that is utilized almost universally to simplify the equations of conservation for natural convection for small temperature differences, including, most importantly, the replacement of $-g - (\partial \rho/\partial x)/\rho$ by $g\beta(T - T_\infty)$, is generally called the Boussinesq transformation, although, according to David Hellums, he should be credited only in part. The approximations of the Boussinesq transformation can be, and increasingly are, avoided in numerical solutions, but at a significant cost computationally. The idealized solutions incorporating the Boussinesq transformation will probably continue to appear in our textbooks and even our handbooks because of their reasonable accuracy and the insight that they provide.

Asymptotic solutions for turbulent free convection Nusselt in 1915 speculated that the local heat transfer coefficient would eventually become independent of distance upward

along a heated vertical plate. Such asymptotic behavior requires the proportionality of the local Nusselt number, Nu_x , to the one-third power of the local Grashof number, $Gr_x = g\rho^2\beta(T_w - T_\infty)x^3/\mu^2$. Fifty- five years later in 1970, I speculated in print that the inertial terms must become negligible relative to the viscous ones as $Pr \rightarrow \infty$ owing to the viscosity, and vice versa, implying proportionality to $Ra^{1/3}$ and $(RaPr)^{1/3}$, respectively. All three of these speculations appear to be validated by the somewhat limited experimental data. The numerically computed values of Shyy-Jong Lin of 1978, which are based on the κ - ϵ model, predict an overshoot (such as that of the orifice coefficient) before approaching the asymptotic proportionality of 1/3, and that probably explains the radically different powers obtained by various experimentalists.

The alternative speculation of Frank-Kamenetskii in 1937 of independence of the asymptotic heat transfer coefficient from both the viscosity and the thermal conductivity, which leads to a proportionality to $Gr^{1/2}Pr$, as well as other speculations that lead to a proportionality to $Gr^{1/3}$ rather than $Ra^{1/3}$, appear to be invalid. As an aside, Eckert and Jackson in 1951 derived, by means of integral-boundary-layer theory, an expression in which Nu_x is proportional to $Gr^{0.4}$. The exponent of 0.4 is simply an artifact of their arbitrary choice of a velocity distribution. One conclusion from this set of analyses is that all speculations need to be tested and a second conclusion is that later speculations are not necessarily better than earlier ones.

Ohm's laws Ohm in 1827 derived expressions for steady-state electrical conduction through resistances in parallel and in series. These expressions are regularly applied for combined thermal conduction and convection and also for surface-catalyzed chemical reactions controlled by mass transfer, adsorption, surface diffusion, and surface reaction in series and parallel (the so-called Hougen-and-Watson expressions).

The radiative heat transfer coefficient If thermal radiation occurs in series or parallel with thermal conduction, forced convection, and/or free convection, it is convenient to linearize the dependence on temperature by means of the following expression in order to allow the application of Ohm's laws, which are restricted to processes that are linear in the potential:

$$h_R \equiv \frac{\epsilon\sigma(T_s^4 - T_\infty^4)}{T_s - T_\infty} = \epsilon\sigma(T_s^4 + T_\infty^2)(T_s + T_\infty) \cong 4\epsilon\sigma T_m^3$$

The exact value of T_m can be calculated for each specified set of values of T_s and T_∞ , but, since the variation of the absolute value is constrained, some arbitrary fixed-mean value is usually employed as an approximation. A *radiative thermal conductivity* may also be defined but it is less useful.

Black-body and gray-body radiation The emission, absorption, reflection, and transmission of thermal radiation are a function of temperature as well as of wavelength for all fluids and solids. An exact treatment taking into account the spectral variations with wavelength is generally so complicated as to be impractical. However, the concepts of black and gray bodies have permitted the derivation of acceptable approximations in many instances, for example, by Bert Larkin in 1959 to characterize the contribution of radiation to heat transfer through insulations, and by John Chen in 1963 for packed beds and later for fluidized beds.

Special Forms of Transport

Fluidized beds As contrasted with the prior listed concepts and variables, those for fluidized beds have, because of its primary development in connection with continuous catalytic cracking, originated almost exclusively within chemical engineering. I will not attempt to summarize the developments in this topic, which continue to be produced to this day, but instead merely mention one early contribution that had a great personal impact. I had

struggled to make sense of experimental data obtained from a pilot plant and then from the startup of the second full-scale fluidized-bed catalytic-cracker, and was overwhelmed by the elementary but perceptive analysis published a few years later in 1948 by Dick Wilhelm and Moosun Kwauk. For example, they noted that

- 1) the pressure drop at the point of fluidization is given by $(-\Delta P)_i = L_i(1 - \epsilon_i)g(\rho_s - \rho)$
- 2) the height of an expanded bed is given by $L(1 - \epsilon) = L_i(1 - \epsilon_i)$
- 3) the mean interstitial velocity can be expressed approximately as $u_{m0} = u_T \epsilon^n$

Packed beds A review of the vast literature on flow through porous media reveals that the majority of the contributions have been by chemical engineers. I will represent this history by a single expression, namely that devised Sabri Ergun, a chemical engineer, in 1952:

$$\frac{-\Delta P}{L} \left(\frac{\epsilon^3}{1 - \epsilon} \right) \frac{D_p}{\rho u_0^2} = 150 \frac{\mu(1 - \epsilon)}{D_p u_0 \rho} + 1.75$$

Although the additive form and the two coefficients of the *Ergun equation* are empirical, the laminar and the inertial (not turbulent) terms both have a theoretical rationale and have proven very successful as compound variables. As an aside, Noel deNevers used the concept of a hydraulic diameter to rationalize the laminar term rather successfully and the concept of a very rough pipe to rationalize the inertial term with less success, perhaps because the latter concept implied turbulent flow.

Laminar condensation Nusselt in 1916 derived the following solution in closed-form for the space-mean heat transfer coefficient for condensation in a laminar film by the ingenious choice of idealizations, namely an isothermal vertical plate of length L and temperature T_p , a pure saturated vapor at T_g , negligible heat capacity, inertia, and fully developed (parabolic) non-rippling flow for the condensate, and negligible drag for the vapor:

$$\Gamma \left(\frac{\mu_\ell \lambda^3}{g \rho_\ell^2 k_\ell^3 (T_g - T_p)^3 L^3} \right)^{1/4} = \frac{4^{3/4}}{3} = 0.9428$$

Here, Γ is the mass rate of condensation per unit breadth of the plate and λ is the latent heat per unit mass.

This solution can be interpreted as an ultimate example of the unique utility of a compound variable. Those of you who are scornful of such a purely theoretical result should ask yourself how much experimentation with what degree of accuracy would be necessary to produce such a relationship, that is lead to the identification of the power-dependence of the rate of condensation on each of these variables. Dimensional analysis would get you part way but not to this degree of resolution. Theoretical and/or numerical solutions have since been derived that avoid each of the idealizations of Nusselt, and they should be used for design. Even so, his solution remains a touchstone both because of the insight provided by its combination of variables and because of the fairly accurate quantitative agreement of its predictions with experimental data.

Some textbooks present this solution rearranged in terms of the Nusselt number (and thereby the heat transfer coefficient) as a function of the Reynolds number, but that is a misdirected effort because the rate of flow (the rate of condensation), which is a dependent rather than an independent variable, is needed to evaluate the Reynolds number.

Several Forms of Transport about Which You May Not Know

Thermoacoustic convection Incorporation of Fourier's law of conduction in the unsteady-state one-dimensional differential energy balance results in

$$\rho c \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right)$$

It has long been recognized by mathematicians that this model is faulty in that it predicts an infinite rate of propagation of energy for some thermal boundary conditions. Cattaneo in 1948, Morse and Feshbach in 1953, and Vernotte in 1958 independently proposed the following so-called *hyperbolic equation of conduction* to avoid that defect:

$$\rho c \frac{\partial T}{\partial t} + \frac{k}{u_T^2} \frac{\partial^2 T}{\partial t^2} = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right)$$

Here, u_T is the velocity of the thermal wave, usually taken to be the velocity of sound. This expression, which they rationalize on the basis of a misreading of the Maxwell's classical paper on the kinetic theory of gases, is pure rubbish with no theoretical or experimental basis, but I have identified over 200 solutions of it in the literature of applied physics and engineering, including a few by chemical engineers whom I will generously grant anonymity. It may be noted that liquids and solids, as well as gases, are sufficiently compressible to result in the generation of thermoacoustic waves.

Meanwhile, back in the realm of rationality – beginning in 1899 with Rayleigh, who postulating the waves to have a sinusoidal shape, and extending to the present, with numerical methods – thermal waves have been predicted with no heuristics by solving the differential energy, momentum, and mass balances as coupled by a variable density. Matthew A. Brown, with my urging, measured the velocity and shape of a wave generated by the sudden and extreme heating of the metallic end-surface of a gas-filled tube. The wave-shape disagreed decisively with all prior predictions, including our own. Upon close examination we detected some evidence of instability in the numerical calculations. It took a reduction of 10^4 in the steps in both space and time (10^8 in all) to eradicate that instability, but we were rewarded with agreement. The applications of thermoacoustic waves, other than thunder as generated by lightning) are mostly rather esoteric but this experience is a valuable reminder of the error and dangerous conclusions that may result from heuristic models, a lack of experimental measurements, misplaced faith in classical models, the misinterpretation of prior work, and/or insufficient tests of numerical computations.

The thermal conductivity of dispersions Maxwell in 1873, using the principle of *invariant imbedding*, derived a solution for the electrical conductivity of dispersions of spheres based on the postulate that the spheres are far enough apart so that deviations in the electric field generated by one sphere do not interfere significantly with those generated by adjacent ones. In 1986 I discovered that his solution, when re-expressed in thermal terms and rearranged in terms of only one compound dependent and one compound independent variable, represented, with minimal parametric dependence, the values determined from the closed-form solution of Rayleigh in 1892 and the numerical solutions of Howard Brenner in 1977, of Andy Acrivos in 1982, and others, for several ordered arrays. Even more remarkably, this solution provides a lower bound and a fair representation even for the extreme of a packed bed and even for granular materials. The corresponding solution for an array of long cylinders with their axes perpendicular to the direction of conduction, such as with fiberglass insulations, is readily formulated. This is a prime example of the utility of an identification of compound variables by means of a theoretical solution for a highly idealized condition.

The migration of water in porous media In order to test predictions of the rate of freezing of the soil outside underground tanks for the storage of liquefied natural gas (LNG) I persuaded Jai P. Gupta to measure, on the laboratory scale, the transient temperature

produced by a flat, cold, bounding surface in sand with a representative concentration of water. He decided on his own to measure the transient concentration as well. As he reported in 1971, the experimental rate of heat transfer through the soil was in good agreement with theoretical predictions but the rate of moisture migration and thereby the rate of freezing was an order of magnitude greater than expected. My colleague David Graves conjectured, and we thereupon confirmed, that this rapid migration occurred in the liquid phase, was a consequence of the variation in surface tension of the water with temperature, and required the presence of a vapor phase. Consequent experiments with heating of the bounding surface produced an even greater surprise, and one with practical consequences in industrial drying, namely the migration of the water in the pore-space toward rather than away from a heated surface. We soon recognized that this reversal in the direction of the migration was a consequence of a maximum in the value of the surface tension of water at 4°C.

What is the description of this experience, which does not appear to involve either modeling or correlation, doing here? The answer is that it was the agreement of the measurements with the theoretical model for diffusion on the one hand and their failure on the other that called attention to their significance. The failure of the experimental results for heating to follow those for cooling had a similar consequence. In this instance, the combination of experimentation and theory produced two discoveries that neither would have done alone.

Similarity Transformations

A major factor in improved understanding and predictions in transport in laminar flow or the absence of flow has been the identification of similarity transformations, that is of combinations of variables that allow reduction of the order of the differential equation(s) of conservation, for example from partial to ordinary. In the dim past most of them were identified one by one and were considered to be signal achievements if not evidence of a stroke of genius. A sampling of familiar ones follows.

Transient thermal conduction Formal substitution of the variable $\xi = kt/\rho c x^2$ for t and x reduces the partial differential equation for transient thermal conduction in a semi-infinite region of an incompressible material with a uniform initial temperature and an imposed surface temperature to an ordinary differential equation that can readily be integrated. The origin of this transformation is unknown to me.

The thin boundary-layer transformation Prandtl in 1904 discovered a similarity transformation that reduced the partial-differential description of developing flow in the “thin laminar boundary regime” on a flat plate to an ordinary differential equation. This transformation led to series and numerical solutions for both flow and convection.

The Pohlhausen transformation E. Pohlhausen in 1921 discovered a similarity transformation that reduced the partial differential model for free convection on a vertical isothermally heated plate, in the thin-laminar-boundary-layer regime, to an ordinary differential model. It led to nearly exact numerical solutions thereof.

The Lévêque transformation Lévêque in 1928 ingeniously approximated the partial-differential model for laminar forced convection in the inlet region of a channel and discovered a similarity transformation that reduced that idealized model to an integral that could readily be solved numerically. The *Lévêque solution* is an invaluable asymptote for the regime near the entrance of a tube or channel, in which the Graetz solution converges too slowly to be useful and in which a finite-difference solution requires excessive discretization.

The Hellums-Churchill methodology In 1964, David Hellums, together with the speaker, devised a simple methodology that can be used to determine whether or not a similarity transformation is possible, and if so to derive it, thereby eliminating the need to be a genius

such as Prandtl, Pohlhausen, and L ev eque. For example, the four aforementioned illustrative similarity transformations are each readily derived by undergraduates after only minimal instruction. Charles W. White, III, a graduate student of Warren Seider, on my urging, wrote a computer program that not only searches for similarity transformations but also performs the otherwise tedious reduction to an ordinary differential equation.

The integral transformation of Saville At my behest, Dudley Saville attempted to carry out a finite-difference solution for laminar free-convection from an unbounded horizontal cylinder of infinite length. He soon discovered that I had chosen an ill-posed problem – the buoyant flow generated by heating or cooling such a hypothetical object sets the whole universe in motion. However, this failure led to a great leap forward. He retreated to the thin-boundary-layer model and discovered a modification of the integral transformation of G ortler that led in 1965 to a common solution for cylinders of general contour in the form of an infinite series that converges more rapidly than the classical solution of Blasius for a round cylinder. He then utilized this methodology to develop the corresponding generalized solutions for horizontal cylinders and vertical axially symmetric bodies of general contour, and then for simultaneous heat and mass transfer.

Conventional Correlating Equations

Although theoretical concepts have had an ever-increasing role in the Century of the AIChE, the design of chemical plants and the analysis of chemical processing has from the beginnings to the present required the use of graphical representations or empirical correlating equations for both physical-chemical properties and for the processes themselves.

Dimensional analysis The correlating equations for transport have generally consisted of the product of powers of dimensionless groupings of dimensional variables. These dimensionless groupings are ordinarily determined by means of applying dimensional analysis to a listing of a sufficient set of variables, including physical properties, to characterize the process. It might appear unnecessary to review this topic, which is presumably well-known to all chemical engineers, but my experience indicates otherwise.

Rayleigh in 1915 got dimensional analysis, as applied to a listing of variables, exactly right. It is my personal opinion that the world would be better off if we simply ignored all subsequent contributions (except possibly as sources of examples). I can't take time today to describe his process in detail and will only note that he postulated an expression in the form of a power series of a term made up of the product of powers of all of the variables. He then focused his attention on only one of these terms and determined these powers insofar as they are constrained by the conservation of dimensions, including time. A typical solution might be

$$Nu = ARe^n Pr^m + BRe^{2n} Pr^{2m} + \dots$$

Rayleigh realized that this result does **not** mean that the Nusselt number is proportional to the product of a power of the Reynolds number and a power of the Prandtl number. Rather it merely indicates that Nu is some unknown arbitrary function of Re and Pr , that is that

$$Nu = \varphi\{Re, Pr\}$$

Unfortunately, many chemical engineers, including academics, fail to make this distinction and then compound the error by plotting experimental data for Nu vs Re and/or Pr on log-log coordinates to determine values of A , n , and m . I wasted a significant amount of time in my younger years trying to rationalize the prevalent value of $n = 0.8$ as in the former expression because I confused it with $4/5$. I eventually realized that meaningful powers of dimensionless groups only occur in asymptotes and that values such as 0.8 are rounded-off artifacts of the choice of some particular range of the variable (here Re) and have no theoretical significance.

If I convinced all or even some of you to abandon correlating equations in the form of arbitrary power functions and their products, as well as the use of log-log plots for their numerical evaluation, this would truly be a Centennial Day for you and for me. I have spent much of the last third of my professional life trying to make that choice possible, and plan to focus some of my remaining few minutes here to try to recruit you to help.

A Correlating Equation for Almost Everything

I am certain the recognition that power-dependences and their products occur only in asymptotes came to many of you before it did to me. However, it came to me as such an epiphany that I vigorously pursued the consequences; one of which was the further realization that exact or nearly asymptotes could be derived for most behavior, not only in transport, and not only in physics, but also in non-technical venues such as athletics and economics. It struck me that incorporating asymptotes in correlating equations might improve their accuracy both numerically and functionally. That proved to be so beyond all expectations. After some trial and error we arrived in 1972 at

$$y_0\{x\} = [y_0\{x\}^p + y_\infty\{x\}^p]^{1/p}$$

I eventually called this expression “the correlating equation for almost everything” in recognition of its universality, but have also referred to it as the *CUE* (for Churchill-Usagi equation) in the interests of brevity and in acknowledgement of my collaborator, Renéé Usagi.

The *CUE* can be seen to be the p th-power-mean of the two limiting asymptotes, $y_0\{x\}$ and $y_\infty\{x\}$. In order to evaluate the combining exponent, p , it is convenient to express the *CUE* in one or the other or both of the following canonical forms;

$$\frac{y\{x\}}{y_0\{x\}} = \left[1 + \left(\frac{y_\infty\{x\}}{y_0\{x\}} \right)^p \right]^{1/p} \quad \text{and} \quad \frac{y\{x\}}{y_\infty\{x\}} = \left[1 + \left(\frac{y_0\{x\}}{y_\infty\{x\}} \right)^p \right]^{1/p}$$

This expression is successful almost beyond belief because it reduces the task of correlation to that of representing the deviation from the closest asymptote rather than the whole variance. I will subsequently mention some examples that illustrate and confirm that assertion.

We were not the first to utilize this expression; earlier users include Andy Acrivos and Tom Hanratty. Our contribution was to recognize its potential for general usage and to devise a standardized procedure for determining the best value of the arbitrary exponent p . Statistical evaluation of the this exponent is unnecessary due to the insensitivity of the predictions to that quantity, and indeed a ratio of two integers may be chosen for convenience

In general the optimal value of p is arbitrary and without theoretical significance, but Eli Ruckenstein derived a theoretical value of 3 for combined free and forced convection, which, happily, was in agreement with the value we had already determined painstakingly from experimental data and numerically computed values.

As an aside, the aforementioned *Ergun equation* can be interpreted as an application of the *CUE* with $p = 1$. Several other examples are mentioned immediately below, and others subsequently in the treatments of analogies, turbulent flow, and turbulent convection.

Restrictions on the use of the CUE

- Asymptotes must be known, derived, or formulated
- Asymptotes must intersect once and only once
- Asymptotes must both be upper or lower bounds
- Asymptotes must be free of singularities

The behavior for large and small values must be symmetrical
 Separate expressions are necessary for derivatives and for integrals, but derivatives
 and integrals of the asymptotes can be utilized

Examples

Laminar free convection The first utilization of the *CUE* in 1972 consisted of the following combination of the asymptotes of LeFevre of 1957 for the local Nusselt number in free convection from an isothermal vertical plate in the thin laminar boundary layer regime:

$$Nu_x^p = \left[0.6004 Gr_x Pr^{1/2} \right]^p + \left[0.5027 (Gr_x Pr)^{1/4} \right]^p$$

The right-most and middle terms are the asymptotic solutions for $Pr \rightarrow \infty$ and 0, respectively. A combining exponent p of 9/4 was found to represent experimental data and numerically computed values of Nu_x for a wide range of values of Pr and Gr_x within 1%.

With this combining exponent, the correlative expression can be reduced to

$$Nu_x = 0.5027 (Gr_x Pr)^{1/4} / [1 + (0.492/Pr)^{9/16}]^{4/9}$$

The denominator of this expression has proven to be such a good approximation for all geometries and thermal boundary conditions, and for the turbulent as well as the thin-laminar-boundary-layer regime, that it has been called “the universal dependence on the Prandtl number”. That is excessive praise; slightly improved representations can be obtained with slightly different coefficients and combining exponents for each condition.

It should be noted that although the goal was a correlating equation for the dependence on the Prandtl number, the dependence on Gr_x was incorporated by virtue of its presence in the asymptotes. Furthermore, insertion of this expression for Nu_f in the previously presented equation of Langmuir provides a fair prediction for Nu_x for values of Gr_x below the range of thin-boundary-layer theory.

The orifice coefficient As an example of the application of the *CUE* of more general interest, we derived in 1974 an expression for the sharp-edged-orifice-coefficient for all values of Re using asymptotes for three regimes. For the asymptote for large Re we initially used 0.611. Empirical asymptotes for the regimes of creeping and transitional flow, as well as numerical values for the two combining exponents were initially derived from the 1933 experimental data of Tuve and Sprenkle for $a_o/a = 0.4$. The resulting predictive equation was subsequently improved by replacing the limiting value of 0.611 by the aforementioned exact value of 0.5793, and the coefficient of 0.16 by the exact value of $1/(12\pi)^{1/2} = 0.1629$ from the solution of Roscoe in 1951 for creeping flow in the limit of $a_o/a \rightarrow 0$, which I belatedly learned about from Al Baer. Presumably, in the intervening years since 1933, the experimental measurements of Tuve and Sprenkle have been improved upon and extended to other values of a_o/a , or have been supplemented or replaced by numerical solutions, but I leave the up-dating to one of you, who may thereby place your name in future handbooks and textbooks.

Multiple variables Multiple independent variables may be taken into account with the *CUE* by either or both of two means. First, secondary variables may be incorporated in either or both of the asymptotes, as was Gr_x in the expression for laminar free convection, or by serial application. If expressions are known from theoretical considerations or can be contrived for consecutive regimes such as creeping, laminar, transitional, and turbulent flow, the application is usually straightforward except for the arbitrary choice of the order of combination. However, in many instances, the transition is sigmoidal in form and unknown. If so, it may be possible to represent the intermediate asymptote by an arbitrary power-function, which may or may not be the tangent through the point of inflection. Our first derivation of

such representation – namely for the variation of the effective viscosity of a pseudoplastic fluid with the shear stress – was not only remarkably successful in a predictive sense but also suggested that the power-dependence of the intermediate asymptote (the Ostwald-deWaele model) may be a mathematical artifact rather than a physical phenomenon.

A generalized expression for transitional behavior In 1974, H.J. Hickman, a graduate student at the University of Minnesota, derived, using the *Laplace transform*, an infinite-series solution for fully developed convection in fully developed laminar flow with heat losses due to radial conduction through the tube wall and external insulation, in series with free convection and thermal radiation to the surroundings. In 2001, I recognized that this solution, after the correction of small numerical errors, could be represented exactly by a highly reduced form of the *CUE*, namely

$$\frac{Nu_J - Nu_T}{Nu - Nu_T} = 1 + \frac{Bi}{Nu_T}$$

Here Nu_J and Nu_T are the internal Nusselt numbers for uniform heating and a uniform wall-temperature, respectively, $Bi = U_e D/k$ is the Biot number, and U_e is the overall heat transfer coefficient for the heat losses, I further recognized that an expression of this general form could be used to represent many other transitional processes, an example of which is presented subsequently in connection with turbulent convection in a tube.

Status and future of the CUE

It goes without saying that graphical correlations are more or less incompatible with computer-aided design, although they remain important to reveal the quality of the representation.

A collateral observation is that most of the scatter observed in conventional log-log plots is due to mis-correlation not poor data.

The *CUE* appears to provide useful replacements for those graphs and power-law representations.

In the past 36 years, my students and other collaborators have developed several hundred predictive or correlative expressions in this form, primarily for transport. I am gratified that several recent books on heat transfer present, almost exclusively, the expressions that we have devised, but am disappointed that so few investigators have followed our lead in developing expressions in this form for their own results. I hope that some of you will be inspired to do so.

Analogies

Many of the analogies in common use by chemical engineers were first conceived by scientists and other brands of engineering. Even so, analogies appear to have a more essential role in chemical engineering, perhaps because of the breadth of our interests and the consequent involvement with processes in which flow, heat transfer, mass transfer, and chemical reactions occur simultaneously. The differential equations of conservation for momentum, energy, and species display some commonalities that suggest analogous behavior but they also display fundamental differences. Most of the analogies now in common use sprung from some ingenious insight rather than by comparison of the equations of conservation. I will start with simple ones, go on to more complex ones, and then describe in some detail the development of a relatively new one.

The equivalent diameter Perhaps the most widely used analogy in practice is that of the equivalent diameter. This concept allows correlations for flow and/or heat transfer in one geometry, usually a round pipe, to be utilized as an approximation for other geometries. The concept requires the choice of some arbitrary methodology to determine the equivalent

diameter. The most common choice is the *hydraulic diameter*, which is defined as four times the cross-sectional area divided by the wetted-perimeter. The *hydraulic diameter* results in an over-prediction of the friction factor for laminar flow in a parallel-plate channel by 50% but a lesser error in turbulent flow and in annuli and rectangular ducts. The *laminar-equivalent diameter*, that is the value that produces the exact value for laminar flow, is somewhat more accurate but requires the availability of a solution or correlation for the latter.

The analogy of MacLeod Bob Rothfus and Carl Monrad determined the conditions that force the velocity distribution in fully developed laminar flow in round tubes and parallel-plate channels to be congruent. Remarkably these conditions are exactly equivalent to expression of the results for round tubes in terms of a^+ and y^+ , and those for parallel-plate channels in terms of y^+ and b^+ . This result is intriguing but of little practical importance. They credit a doctoral student, Alexander MacLeod, with the conjecture that this congruence might carry over to turbulent flow. A plot of experimental data by Glen Whan and Bob Rothfus confirmed that conjecture and also demonstrated that it does not apply to the regime of transition. This discovery of congruence is of great importance not only because it allows experimental data for the two geometries to be used interchangeably but even more importantly because it allows the numerical values obtained for parallel-plate channels by *DNS* to be applied for round tubes.

The analogy between heat and mass transfer The analogy between heat and mass transfer is based on the similarity of the equations of conservation. Insofar as they are identical in form, the Sherwood number and the Schmidt numbers can simply be substituted for the Nusselt number and the Prandtl number, respectively, in a correlation or solution for heat transfer. That is useful because the correlations and solutions for heat transfer are much more extensive. However, the similarity may be spoiled by a number of factors, principally the bulk motion generated by non-equimolar mass transfer, the dependence of the Schmidt number on composition, and the differing dependences of the Schmidt and Prandtl numbers on temperature and pressure. It may be noted as an aside that in 1997, Dimitrios Papavassiliou and Tom Hanratty explained, using *DNS*, the failure of the analogy for large values of Sc .

The analogy between electrical and thermal conduction The analogy between electrical and thermal conduction is exact for steady state processes but doesn't apply to unsteady state ones. It has few active applications but two are of great importance. Maxwell's law of dispersions and Ohm's law were conceived in electrical terms but their analogues in chemical and thermal processing have proven to be of great utility.

An analogy for buoyant processes Emmons in 1954, recognizing that laminar free convection, film condensation, film boiling, and film melting are all controlled by thermal conduction across a film moving at a velocity controlled by gravity, derived the following generalized solution:

$$\frac{h_m L}{k} = \left(\frac{\rho F L^3}{K \mu k \alpha \Delta T} \right)^{1/4}$$

Here, F is the gravitational force per unit volume of the film, K is a numerical factor characterizing the viscous shear, and α is the increase in the mass rate of flow of the film per unit of heat transfer. He tabulated values of the factors F , K , and α for each of the four processes on the basis of qualitative considerations. One merit of this analogy is that it allows a common plot of experimental data for all four processes. More accurate coefficients have been derived for each of the four individual processes so the inaccuracy of the tabulated values of Emmons represents an intentional sacrifice to obtain commonality and insight. As

an aside, the term in brackets, after substitution for F , K , and α , may be identified as the Rayleigh number in the case of free convection but the counterparts for the other three gravitational processes do not appear to have an accepted "name".

The corresponding analogy for the turbulent regime is

$$\frac{h_m L}{k} = A \left(\frac{\rho F L^3}{K \mu k \alpha \Delta T} \right)^{1/3}$$

Here, A is an empirical coefficient. Emmons' values of F , K , and α convert this expression to the accepted form for turbulent free convection, but the limited and very scattered experimental data for film condensation, film boiling, and film melting in the turbulent regime do not appear to confirm the predicted independence from L .

In a qualitative if not a quantitative sense, the analogy of Emmons represents one of the broadest generalities in all of transport.

An analogy between chemical reaction and convection Although energetic chemical reactions have been known for over 40 years to enhance or attenuate convection radically this important aspect of behavior does not appear to have found its way into any textbooks or handbooks on heat transfer or mass transfer. The earliest studies are by well-known chemical engineers including P.L.T. Brian, Bob Reid, and Samuel Bodman in the period 1961-1965, Joe Smith in 1966, and Louis Edwards and Robert Furgason in 1968. My encounter with this phenomenon in combustion beginning in 1972, and the absence of a simple predictive expression, eventually led me to derive one. First, I derived for a very idealized set of conditions

$$Nu = Nu_o / (1 + \beta Q)$$

Here Nu_o is for no reaction, Q is the ratio of the heat of reaction to the heat flux from the wall, and β is an arbitrary coefficient. This expression led by a path too long and complex to describe here to the following one relating the local mixed-mean rate of reaction, as represented by $\xi(1 - Z_{mx})$, to the local rate of heat transfer from the wall, as represented by Nu_x :

$$Nu_x = \frac{Nu_{ox}}{1 + \beta \xi (1 - Z_{mx}) \exp \left\{ \frac{E / RT_0}{1 + (\tau [Z_{mx} + (K_{ox} / \xi)])^{-1}} \right\}}$$

This expression, which is restricted to fully developed laminar or turbulent flow with a uniform heat flux on the wall and a first-order reaction, explains as well as predicts almost exactly the the extreme and chaotic enhancement and suppression of the Nusselt number. It differs fundamentally from conventional analogies in that it is for a developing rather than a fully developed process.

Turbulent Flow

I have three reasons for giving this topic more attention than any other. First, most chemical processing takes place in turbulent flow, second, turbulent flow and convection best illustrate the advance of our technical capability over the past 100+ years, third, chemical engineers have had a critical role in its evolution, and fourth, the state of the art in dealing with turbulent flow may not be known to all of you. Most of the topics that follow qualify for the prior section on concepts or that on analogies. They were deferred to this section in the interests of continuity.

Time-averaging In my view, the greatest advance of all time in modeling turbulent flow and transport in turbulent flow was by Osborne Reynolds, who in 1895 space-averaged the partial differential equations of conservation. The equivalent of this space-averaging is now carried

out by time-averaging. Not everyone agrees with my assessment because the process produces some new effective variables such as $\overline{\rho u'v'}$ and $\overline{\rho cv'T'}$, for which arbitrary predictive expressions are required. Even so, all turbulent modeling for nearly a century started with these time-averaged expressions and they may still have some applicability in the future.

The eddy diffusivity In 1877, (and thus before the turbulent shear stress, heat flux density, and mass flux density were defined in terms of the fluctuating components of the velocity, temperature, and composition by Reynolds) Boussinesq introduced the eddy viscosity, eddy conductivity, and the eddy diffusivity to model them. They could have been examined in the section on analogies because Boussinesq conceived of these expressions for turbulent transport as analogs of those for molecular diffusion. I will try to summarize as briefly as possible the path that led us from Reynolds and Boussinesq to where we are today. That is not easy because it involves as many missteps and side-steps as advances.

The power-law for the velocity distribution In 1913, Blasius, a student of Prandtl, fitted a log-log plot of the available data for the friction factor versus the Reynolds number in a round pipe with a straight line of slope of $-1/4$. Prandtl presumed this nice round fraction had some significance, and in 1921 derived from it, with extraordinary ingenuity, a one-seventh-power dependence for the velocity on the radius. He subsequently realized, in part due to the obvious deficiencies of the one-seventh power velocity distribution, that the slope $-1/4$ was merely an artifact of the narrow range of the experimental data available to Blasius.

The mixing-length Having found the power-law to be deficient, Prandtl in 1925 next proposed to represent the turbulent shear stress with $\rho \ell^2 (du/dy)^2$, where ℓ is a *mixing-length* for the turbulent eddies analogous to the mean-free-path of gaseous molecules. Bird, Stewart, and Lightfoot, many years later in 1960, but while this concept was still in vogue, correctly criticized it as a “very poor analogy”. Prandtl conjectured that the mixing length might vary linearly with distance from the wall near the wall, that is, $\ell = ky$, and used this conjecture, along with several clever idealizations to derive

$$u^+ = A + \frac{1}{k} \ln\{y^+\}$$

Because of its obvious failure at the wall and at the centerline, the region of applicability of this expression has become known as the “turbulent core near the wall”. Because of his subsequent, presumably independent derivation of the same expression, the empirical coefficient of linearity, k , is generally known today as the *von Kármán constant*. Von Kármán and Prandtl both further conjectured that, in spite of its failures near the wall and near the centerline, the integration of this expression over the cross-section might yield a good approximation for the mixed-mean velocity and thereby the friction factor, namely

$$u_m^+ = \frac{u_m}{\sqrt{\rho \tau_w}} = \sqrt{\frac{2}{f}} = A - \frac{3}{2k} + \frac{1}{k} \ln\{a^+\} = B + \frac{1}{k} \ln\{a^+\}$$

This derivation is one of the most fateful in history because the resulting expression has by means of a variety of empirical values of A (or B) and k , remained in our handbooks, either algebraically or graphically, usually without the inclusion of supporting data, to this day.

Prandtl in 1925 further conjectured that the mixing length might approach a constant value at the centerline leading to the equivalent of the following erroneous asymptote for that region:

$$u = u_c - Cr^{3/2}$$

In order to encompass a wider range of behavior, von Kármán in 1930 proposed the following theoretical-looking expression:

$$\ell = k \left(\frac{du/dy}{d^2u/dy^2} \right)$$

I once asked him, over a glass of champagne, the source of this, to me, mysterious relationship. Without batting an eye, he replied that it was the simplest expression, involving only derivatives of the velocity, with the correct overall dimension. After considerable analysis, I satisfied myself that this was true. Unfortunately, however, the predictions of this elegant expression are quite erroneous and misleading because of the failure of the very concept of a mixing length.

Wall-based variables In 1926, the year after proposing the mixing length, Prandtl took another tack. First he used routine dimensional analysis to derive

$$u\sqrt{\rho/\tau_w} = \varphi\{y\sqrt{\rho\tau_w}/\mu, a\sqrt{\rho\tau_w}/\mu\}$$

Then he introduced the following symbols which have remained in use for over 80 years

$$u^+ = \varphi\{y^+, a^+\}$$

The law of the wall Prandtl next conjectured that near the wall the dependence on a^+ would phase out leading to

$$u^+ = \varphi\{y^+\}$$

which is known as “the universal law of the wall”. This is a seemingly straightforward application of dimensional and asymptotic analysis but it is not. Had the pressure gradient rather than the shear stress on the wall, or the radius rather than the distance from the wall been chosen as variables, or had some other grouping such as uap/μ been chosen as the dependent variable, the elimination of a would not have led to such a useful result. Although he did not say so in print, my guess is that Prandtl tested and then rejected each of these alternatives as inferior.

The law of the centerline Postulating that the velocity field near the centerline to be independent of the viscosity next lead Prandtl to the following counterpart for this region:

$$u_c^+ - u^+ = \varphi\{a/y\}$$

The term $u_c^+ - u^+$ is called the *velocity defect*.

The law of the turbulent core Millikan in 1938 ingeniously recognized that the only expression that conformed to both of these two functional asymptotes was

$$u^+ = B + \frac{1}{k} \ln\{y^+\}$$

This derivation, which is free of any heurism, reveals that two erroneous concepts (the mixing length and its linear variation near the wall) may lead to a valid result.

The limiting behavior at the wall Prandtl also postulated that very, very near the wall the shear stress due to the turbulent fluctuations and the effect of curvature would be negligible leading to

$$u^+ = y^+$$

This expression can be noted to conform to “the law of the wall.”

The κ - ε model The best-known predictive equation for the eddy viscosity is the κ - ε model of Kolmogorov, Prandtl, and Batchelor. The *kinetic energy of turbulence* κ and the *rate of dissipation of turbulence* ε have physical significance but the equations devised beginning in 1972 by Launder and Spalding and others for their prediction are very empirical, based on homogeneous turbulence, and not very accurate functionally or numerically, particularly near

the wall where the predictions are most important. This model has been superseded for fully developed flow but remain the principal, if unreliable, source for developing flow.

Direct numerical simulation (DNS) Prior to its accomplishment, I conjectured that numerical integration of the time-dependent equations of conservation for the turbulent regime was not feasible because the scale of the turbulent eddies extends from that of the size of the channel down to that of the molecules, and that the grid-size would need to extend down to the latter level. My reasoning was faulty because I failed to realize that the grid-size only needs to be decreased to the point at which the turbulent shear stress is negligible compared to the viscous shear stress. In an informal conversation at the AIChE Annual meeting in 1990 I learned from Charlie Sleicher and Tom Hanratty that they and their doctoral students had followed the lead of Kim, Moin and Moser in 1987 and carried out such calculations, and that they were essentially free from empiricism. I immediately jumped to an even more erroneous conclusion, namely that we had entered a new era in which such calculations would replace all other approaches to turbulent flow and convection. However, after more than 20 years, *DNS* calculations are essentially limited to planar flows at rates barely above that for fully developed turbulence.

Large-eddy simulation (LES) This methodology, as devised by Schumann in 1975, relaxes the restriction on the rate of flow by utilizing *DNS* only for the fully turbulent core, but requires the use of the κ - ϵ model with arbitrary wall-functions or the equivalent for the region near the wall.

We sorely need a new algorithm or concept that will deliver the predictions of turbulent flow promised but not produced by *DNS* and *LES*.

The turbulent shear stress very near the wall In 1932, Eger Murphree, a chemist, and somewhat later, Charlie Wilkie, a chemical engineer, as well as others, proposed on the basis of speculative asymptotic analysis of the time-averaged equation for the conservation of momentum that very near the wall the expression

$$-\rho \overline{u'v'} = \alpha(y^+)^3 + \beta(y^+)^4 + \dots$$

The existence or non-existence of the term in $(y^+)^3$ was disputed for over 50 years because of the difficulty of carrying out experimental measurements of the required accuracy. This third-power dependence is of critical importance because it implies a proportionality of Nu to $Pr^{1/3}$ rather than $Pr^{1/4}$ in the limit of $Pr \rightarrow \infty$. The issue was finally settled definitively in favor of $(y^+)^3$ by the earliest results of *direct numerical simulation (DNS)*, including those of Rutledge and Sleicher and of Lyons, Hanratty, and McLaughlin. This result implies that the velocity distribution near the wall is represented by

$$u^+ = y^+ - \frac{\alpha(y^+)^4}{4} + \dots$$

The afore-mentioned calculations also provided the first reasonably accurate value for the coefficient α , namely 0.00070.

The local fraction of the shear stress due turbulence As an alternative to the representation of the turbulent shear stress by heuristics such as the eddy viscosity and the mixing length, Christina Chan and I in 1995 proposed to represent it directly – at first in terms of $(\overline{u'v'})^+ \equiv -\rho(\overline{u'v'})/\tau_w$, but later in terms of $(\overline{u'v'})^{++} \equiv -\rho(\overline{u'v'})/\tau$. The latter has the advantage

of being finite at the centerline. From their definitions, $\frac{\mu_t}{\mu} = \frac{(\overline{u'v'})^{++}}{1 - (\overline{u'v'})^{++}}$. This result confirms

that, despite its heuristic origin and thereby the contempt of many “purists”, the eddy viscosity really has physical significance, that is, the turbulent shear stress is proportional to the

velocity gradient. (Boussinesq was either lucky or very insightful.) At the same time, the eddy viscosity is inferior in terms of simplicity and singularities to $(\overline{u'v'})^{++}$ and is therefore now of historical interest only. If you are still using it as a variable for fully developed flow you are out of date.

Expressing the mixing length in terms of $(\overline{u'v'})^{++}$ reveals that, despite all the criticism it too is independent of its mechanistic and heuristic origin. However, it is also revealed to be unbounded at the centerline of a round tube or the central plane of a parallel plate channel. How did such an anomaly escape attention for more than 70 years. One explanation is the uncritical acceptance by Prandtl of the plot of values of the mixing length obtained from the “adjusted” experimental values of Nikuradse, followed by the uncritical extension of respect for Prandtl and von Kármán to all of their derivations.

An algebraic correlating equation for the turbulent shear stress Eventually in 2000 we devised, using the CUE, the following expression for the local fraction of the total shear stress due to turbulence:

$$(\overline{u'v'})^{++} = \left[\left[0.7 \left(\frac{y^+}{10} \right)^3 \right]^{-8/7} + \left| \exp \left\{ \frac{-1}{0.436 y^+} \right\} - \frac{1}{0.436 a^+} \left(1 + \frac{6.95 y^+}{a^+} \right) \right|^{-8/7} \right]^{-7/8}$$

The term inside the square brackets is the asymptote for $y^+ \rightarrow 0$, and that inside the absolute value signs that for $y^+ \rightarrow a^+$. An expression for the extreme limiting behavior of $(\overline{u'v'})^{++}$ for that condition was derived from the observed behavior, but was subsequently recognized to have the same form as one derived by Hinze in 1963 on the basis of the observation of a finite value of the eddy viscosity at the centerline. The numerical coefficients and the exponent of $-8/7$ are based on the experimental data of Zagarola. (Note the value of 0.436 for the *von Kármán constant*.) According to the aforementioned analogy of McLeod, this expression for $(\overline{u'v'})^{++}$ is applicable for parallel-plate channels if b^+ is substituted for a^+ . We have also adapted it for circular concentric annuli.

The ultimate correlating and predictive equation for the friction factor Essentially exact expressions for the local and mixed-mean velocities follow directly from that for $(\overline{u'v'})^{++}$. First, the differential momentum balance is integrated to obtain values for $u^+\{y^+, a^+\}$ and $u_m^+\{a^+\}$ in terms of $(\overline{u'v'})^{++}$. These values are then used to evaluate the coefficients and combining exponents for correlating equations constructed from integrals of the asymptotic terms of the correlating equation for $(\overline{u'v'})^{++}$. The resulting expression for u_m^+ and thereby for the friction factor in a round tube is

$$u_m^+ = \sqrt{\frac{2}{f}} = 3.3 - \frac{227}{a^+} + \left(\frac{50}{a^+} \right)^2 + \frac{1}{0.436} \ln \left\{ \frac{a^+}{1 + 0.301(e/a^+)a^+} \right\}$$

The resulting expressions for the local turbulent shear stress and the local time-mean velocity, as well as this one for the mixed-mean velocity (equivalent to the friction factor), represent the experimental data and the essentially exact numerically computed values for these quantities within their uncertainty. The unfamiliar terms in $227/a^+$ and $(50/a^+)^2$ arise from the boundary layer, which has been neglected explicitly in all prior integral expressions.

An iterative solution is required to determine the friction factor for a specified value of $Re = 2a^+ u_m^+$ but the convergence is rapid, and the gain in accuracy, relative to expressions

explicit in Re , more than justifies the necessity of the iteration. (This conclusion of mine is regularly contested, most recently in the March 2008 issue of *Chemical Engineering Progress*, but I am unconvinced that iteration is unacceptable or even onerous.)

The following expression in the form of two consecutive applications of the *CUE* predicts the friction factor for a round tube for all regimes of flow, including transitional, and all effective roughness ratios:

$$f = (f_l^{12} + [f_t^{-16} + f_T^{-16}]^{-3/2})^{1/12}$$

Here, $f_l = 16/Re$ (Poiseuille's law), $f_t = (Re/37530)^2$, and f_T is the above expression for fully turbulent flow. This expression is a complete replacement for and improvement on all expressions and plots for the friction factor. Although it obviates the need for one, it can readily be programmed to produce such a plot in every detail.

Experimental Data

The most coherent and comprehensive set of experimental data in the history of transport is that of Nikuradse in 1930, 1932, and 1933 for the velocity distribution and pressure drop in flow through piping. He recognized that the failure of his measurements to conform to the theoretical prediction by Prandtl of $u^+ \rightarrow y^+$ for $y^+ \rightarrow 0$ must be due to experimental error. Unfortunately, rather than repeat his measurements with improved instrumentation he "adjusted" the values to fit the theory of his doctoral advisor. Had Nikuradse not done so, Prandtl would presumably recognized the fundamental failure of the concept of a mixing length 40 years before Christina Chan and I proved it to be fundamentally in error in even a mathematical sense.

Although many measurements of the velocity distribution and pressure gradient were made through the years, a comprehensive set of data to replace these "adjusted" values outright had to wait for Zagarola in 1996, and his were only for smooth pipe and did not extend quite to the lower limit of the turbulent regime. Despite heroic efforts by Zagarola and his colleagues in the Astronomy department at Princeton University to polish the surface, I conclude that even at the highest Reynolds numbers he attained,, the roughness of his tubing affected the measurements.

Turbulent Convection

As contrasted with turbulent flow, whose exiguity I have recounted in terms of asymptotics and speculations, the history of turbulent convection unfolds almost wholly through analogies between momentum and energy transfer. Literally dozens of such analogies have been proposed, most incorporating some novelty and/or minor advancement. I have chosen a limited, arbitrary set of those that have served as primary quantitative predictors of turbulent convection. However, before turning to the analogies, one solution and one correlating equation, and several quantities and relationships need to be identified.

The solution of Sleicher As most of you know, Graetz in 1883 devised an infinite series solution for developing convection in fully developed laminar flow following a step in wall temperature. Despite its limitations in a practical sense, the Graetz solution is one of the icons of thermal science. Charles Sleicher in 1956 devised the equivalent series for turbulent flow and convection using empirical correlating equations for the velocity distribution and eddy diffusivities, and an analog computer to determine the eigenvalues and eigenfunctions. In 1969, Notter and Sleicher greatly improved that accuracy of that solution using a digital computer and updated expressions for the velocity distributions and eddy diffusivities.

Generalized correlating equations for forced convection In 1977, I devised, using the *CUE* with 5 asymptotes and four combining exponents, a correlating equation for Nu and Sh for all Pr and Sc , respectively, and all Re (including the laminar, transitional and turbulent

regimes). The same structure, but with different coefficients, was proposed for uniform heating and uniform wall-temperature. A graphical comparison of these expressions with experimental data and a few numerically computed values of Sleicher and Notter revealed that, except for mass transfer for very large values of Sc , the discrepancies are generally within the uncertainty of the data. Those discrepancies are now presumed to be due to a breakdown of the analogy between heat and mass transfer. Complementary expressions were also devised for developing convection. The asymptotes for Nu and f for the turbulent regime have been superseded by the afore-presented improved expressions, but the overall structure and the overall correlating equation remain valid.

The eddy conductivity and the turbulent Prandtl number The turbulent conductivity ratio is analogous to the eddy viscosity ratio, and the fraction of the local heat flux density due to the turbulent eddies, namely, $(\overline{T'v'})^{++} \equiv \rho c_p (\overline{T'v'}) / j$, to $(\overline{u'v'})^{++}$. It proves convenient to introduce still another analogous variable, namely the turbulent Prandtl number ratio,

$$\frac{Pr_t}{Pr} = \frac{(\overline{u'v'})^{++} [1 - (\overline{T'v'})^{++}]}{(\overline{T'v'})^{++} [1 - (\overline{u'v'})^{++}]}$$

in place of $(\overline{T'v'})^{++}$ in the differential energy balance. Although Pr_t varies with Pr and with $(\overline{u'v'})^{++}$, its variation is more constrained than that of $(\overline{T'v'})^{++}$.

Peter Abbrecht in 1956 determined the eddy conductivity experimentally in a developing temperature field and thereby confirmed his own conjecture that it was independent of the temperature field and thereby of the thermal boundary condition. It follows that Pr_t/Pr and $(\overline{T'v'})^{++}$ are as well. Furthermore, from the analogy of MacLeod it can be inferred that k_t/k , $(\overline{T'v'})^{++}$, and Pr_t/Pr , are identical for a round tube and a parallel plate channel when expressed in terms of a^+ and b^+ , respectively. The limited experimental data appear to confirm this conjecture.

The Reynolds analogy Osborne Reynolds in 1874 postulated that momentum and energy were transported at equal mass rates from the bulk of the fluid to the wall by the oscillatory radial motion of turbulent eddies and thereby obtained a result that can be expressed in modern terms for a round tube as

$$Nu = Pr Re f / 2$$

or in terms of individual dimensional variables as

$$h = 2\tau_w c / u_m$$

The latter expression reveals that the postulates of Reynolds imply independence from the diameter as well as from the viscosity, density, and thermal conductivity. The *Reynolds analogy* is noteworthy, not only for its date of appearance and its freedom from any specific mechanism of turbulent transport, but most importantly for its semi-quantitatively correct predictions. Remarkably, it remains implicit in the latest expressions for turbulent convection.

The Prandtl-Taylor analogy Prandtl in 1910 and G.I. Taylor, independently in 1916, attempted to correct for the major conceptual shortcoming of the Reynolds analogy by postulating that the eddies penetrate only to a finite distance from the wall, δ_s , called the *laminar sublayer thickness*, and that the transport of momentum and energy across that distance occurs by molecular diffusion. Their solutions were for unconfined flow along a flat plate but they can be rewritten approximately as follows for a round tube:

$$Nu = \frac{Pr Re f / 2}{1 + \delta_s^+ (Pr - 1) (f / 2)^{1/2}}$$

Max Jakob in 1949 remarked that “There exists a whole literature about (the equivalent of δ_s^+ .)” However, even a fixed numerical value of say 11 provides a reasonable prediction of Nu for all values of Re . Although the *Prandtl-Taylor* analogy has the merit of correctly predicting a shifting proportionality of Nu from $Re/2$ to $Re(f/2)^{1/2}$ as Pr increases, it shares the shortcomings of the Reynolds analogy for $Pr \leq 1$, and erroneously predicts independence from Pr for $Pr \rightarrow \infty$.

The Reichardt analogy Reichardt in 1951 developed a greatly improved analogy by dividing the one-dimensional differential momentum balance in terms of the eddy viscosity ratio by the equivalent differential energy balance, thereby eliminating y^+ , the dimensionless distance from the wall. He then introduced a potpourri of ingenious physical and algebraic approximations that allowed him to integrate the resulting expression and obtain a closed-form solution for Nu in terms of Re , Pr , and f . The *Reichardt* analogy, although far more accurate functionally and numerically than that of Prandtl and Taylor, has not received much direct usage because it is also far more complicated, requiring empirical graphical representations for its several variable coefficients. Its structure has however been utilized in most subsequent analogies, including those of Bill Friend and Art Metzner in 1958, Petukhov in 1970, Volker Gnielinski in 1976, and my own in 1997.

A reinterpretation and improvement of the Reichardt analogy Churchill, Shinoda, and Arai in 2000 noted that the Reichardt analogy could be interpreted as an interpolating equation in the form of the *CUE*, thereby greatly simplifying its expression and its implementation. Churchill and Zajic in 2001 took advantage of this reinterpretation to devise greatly improved analogies for all values of Pr and Re . As an example, their expression for $Pr \geq Pr_t$ is

$$\frac{1}{Nu} = \left(\frac{Pr_t}{Pr}\right) \frac{1}{Nu_1} + \left[1 - \left(\frac{Pr_t}{Pr}\right)^{2/3}\right] \frac{1}{Nu_\infty}$$

Here, $Nu_1 = \beta Re/2$ and $Nu_\infty = 0.07343 \left(\frac{Pr}{Pr_t}\right)^{1/3} Re \left(\frac{f}{2}\right)^{1/2}$. The presence of the very first analogy

(that of Reynolds) in this latest analogy should be noted. The coefficient β is an exactly defined function of the velocity distribution but the expressions representing this functionality, which differ for uniform heating and uniform wall-temperature, are too complicated to include here. The coefficient of Nu_∞ , namely $3^{3/2}(0.0007)^{1/3}/2\pi = 0.07343$, is empirical only by virtue of its incorporation of the coefficient of 0.0007 in the previously given expression for $(\overline{u'v'})^{++}$ very near the wall.

The Colburn analogy The best-known analogy between momentum and energy transfer to chemical engineers is that derived by Alan Colburn in 1933. As contrasted with all of those I have just described, it has no mechanistic or theoretical basis. With great insight he observed a possible functional and numerical similarity between the following empirical correlating equations of E.C. Koo, a doctoral student at MIT, for the friction factor, and of Dittus and Boelter for the Nusselt number:

$$f = 0.046/Re^{0.2}$$

$$Nu = A Re^{0.8} Pr^n$$

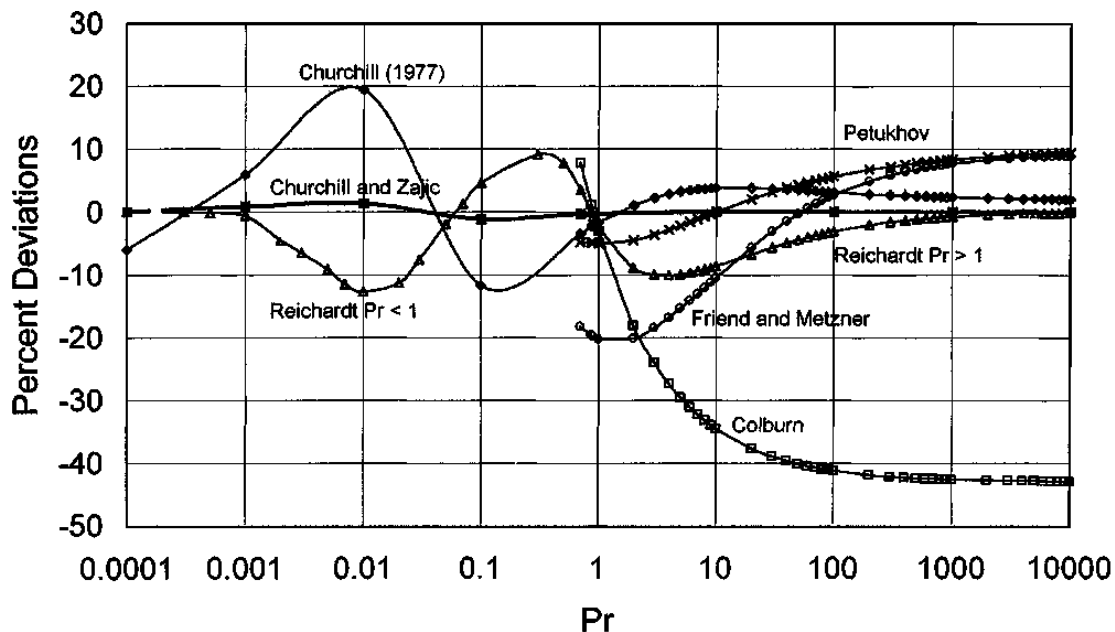
The recommended values of A and n were 0.0243 and 0.4 for heating the fluid, and 0.0265 and 0.3 for cooling it. (As an aside, the gross functional inadequacy of the Dittus-Boelter equation was first exposed by its failure to predict heat transfer for liquid metals when they came into widespread use in connection with nuclear reactors in the 1950s.)

Colburn recognized that taking the ratio of these two equations would result in one in which Nu was proportional to Re^f . In order to force the net coefficient to be an integer, he chose $A = 0.023$ for both heating and cooling, which was quite a stretch, and as a compromise between 0.4 and 0.3 chose $n = 1/3$. He later disclaimed any theoretical basis for that choice. The final result was

$$f/2 = Nu/RePr^{1/3}$$

He named the grouping on the right-hand side the j -factor, because of its analogy to the f -factor (friction factor), giving it a life of its own. This expression, together with an empirical correlating equation for the friction factor, remains in use to this day, although it is seriously wrong functionally in every respect, and, as I will now show you, numerically as well.

A graphical comparison of analogies for turbulent convection



This plot of the percent error of the various predictive expressions for turbulent convection, as compared with essentially exact numerically computed values, reveals that the Colburn analogy is grossly in error except for $Pr = 1$, and that all of the analogies except that of Zajic and myself are in modest error for most values of Pr . Although this plot is for $Re = 227,000$ ($a^+ = 5000$), we obtained similar results were for all values of Re above 4000.

I have asked a number of experts in process heat transfer if they were aware that the predictions of the Colburn analogy were in such error. The most common answer is that they know the predictions of their computer package for heat transfer are grossly low, and that they compensate by applying a personal safety factor. I believe it is not disrespectful to Alan Colburn, whom I knew and greatly admired, to suggest the discard of a relationship that has out-lived its usefulness.

Devising Algorithms

Without stepping on the toes of Bruce Finlayson, I would like to mention some contributions of chemical engineers to numerical computations that I presume to be outside the scope of his talk. Advances in computer hardware and software, including the development of computational algorithms for simulation, are not the whole story. Conceptual advances as primed by need have played a significant role in the advancement of transport. A few examples follow.

Electronic computational machinery first became available at the very time that I began my professorial career in 1952, and I subsequently have asked all of my doctoral students to undertake numerical solutions as well as closed-form solutions and experimental measurements. The state of the computational art was not only primitive when we began, but, despite all the formal advances, has to this day lagged behind their needs, forcing them to devise their own numerical algorithms. As an illustration of the ingenuity of chemical engineers in this respect, I will first mention a few such contributions primarily in just one narrow form of transport, namely natural convection in enclosures.

In 1952, when a "large-scale" electronic digital computer became available to us, William R. Martini asked if I believed that the partial differential equations of conservation described the real world and if we solved them numerically would we obtain accurate descriptions thereof. With some bravado, I replied "Certainly", leading him to attempt the first-ever two-dimensional computations for confined natural convection. He had only semi-quantitative success because of limitations in hardware, but, as a result of his fortuitous use of an unsteady state formulation he discovered that the process was oscillatory even before it was revealed to be so by his experimental measurements.

Using conventional algorithms, David Hellums in 1960 succeeded in obtaining convergent solutions that matched Martini's quasi-one-dimensional but oscillatory experimental results.

Jim Wilkes in 1963 extended the scope of the computations to predict the transient, truly two-dimensional motion in a rectangular channel by introducing the stream function and the vorticity as variables.

Mike Samuels in 1967 extended these computations to predict the criteria for instability by introducing a "false-transient" term for the stream-function equation, thereby rendering it parabolic.

In the same year Khalid Aziz and David Hellums at Rice University carried out the first three-dimensional calculations for confined natural convection by utilizing the representation of George Hirasaki in terms of "vector potential".

Humbert H.-S. Chu in 1976 calculated the effects of heater-size and location on the energy requirement and temperature distribution in living or working space with losses to the surroundings, and obtained surprising results of great practical interest. At first, he appeared to have paid a penalty for the unorthodox use of a non-conservative finite-difference formulation, namely a different heat flux entering and leaving the space. However, he then made an important discovery, namely that the difference between the computed values of the entering and exiting fluxes was a far better measure of the rate of convergence than the conventional ones.

In the same year, Hiroyuki Ozoe and his co-workers at Okayama University introduced the isometric representation of particle paths to describe the fluid motion in three-dimensional natural convection as an analog of the stream function in two dimensions and discovered that all paths are concentric double helices, or degeneracies thereof. They confirmed this behavior predicted experimentally.

In 1982, Paul P.-K. Chao developed a program for displaying particle paths in real time long before standard algorithms for this purpose became available.

In 1995, Vicki B. Booker and coworkers at the NEC Corporation in Tsukuba, where she was a visitor, found experimentally that Czochralski crystallization, which is widely used to make pure silicon, is oscillatory, thereby rendering her steady state computations, made at the University of Pennsylvania, invalid. She confirmed this observation computationally by

formulating an unsteady-state algorithm that predicted transient behavior but whose long-term execution required computational times beyond that available.

In 1998, Hiroyuki Ozoë and coworkers at Kyushu University subsequently confirmed the existence of oscillations in confined natural convection in low-Prandtl-number fluids even for the simpler geometry and boundary condition of parallel horizontal parallel plates heated from below (*the Bénard problem*). The circulation shifts erratically between hexagonal cells and long cylindrical roll cells.

In 1971, Warren Seider, in the process of numerically modeling the developing flow and transport following the merging of a central jet and an outer annular flow, unexpectedly encountered an instability that was subsequently confirmed experimentally. The computational instability was found to be attributable to the finite thickness of the short inner tube, while, on the other hand, the experimental instability was found to be due to any minute radial perturbation in the flow, such as that caused by a bubble of air on the tube wall or someone walking in the laboratory. He ingeniously avoided the numerical instability by carrying out the computations in time, starting from a condition of no flow, thereby confirming experiments in which all physical perturbations were minimized and the flow remained stable as it developed.

The Marker-and-Cell model of Harlow and coworkers is uniquely applicable for solving the partial differential equations of conservation numerically for unbounded flows. Eddy A. Hasbun, in 1973, utilized it successfully to predict the extrusion of Plexiglas^R in three dimensions.

The lesson from these examples is that the prediction or simulation of transport often depends on conceptual innovation, either mathematically or physically.

Simulation

Simulation in the current sense allows us to use correlations for transport to predict complex behavior for the purposes of design and analysis. The advancement and current state of simulation are beyond the scope of the current presentation. However, it seems appropriate to note that this process invokes a hidden risk, namely the possible error due to out-of-date and erroneous correlating equations imbedded in computer packages. Simulation has another almost contradictory role that has been implicit in this presentation, namely the prediction of detailed behavior from “first principles” in order to produce “computed values” to supplement experimental data in support of the construction of correlating equations.

The Influence of AIChE Programming

Industrial and academic attendees have a basic conflict over programming at AIChE meetings. Academics generally prefer to present work in progress with the hoped that that the presentation will provoke constructive criticism and perhaps establish informal proprietary rights. They are inclined to entrust the presentation to a graduate student because of the invaluable professional experience, the start in networking, and the encouragement to participate in future meetings. Accordingly they resist the burden of preparing and distributing a document that will soon be out of date. Academic attendees who are not making presentations understand and support this posture.

On the other hand most industrial participants attend the technical presentations hoping to learn something that will help with their current project and appreciate a manuscript to take back to their work place as evidence of their attendance and as proof that it was worth the time and cost. They also would favor a presentation by the faculty member who supervised the work rather than by the graduate student who actually did most of the work. .

In 1970, the Program Committee, hoping to increase industrial attendance promulgated the policy “No paper, no podium.” A group of “young Turks” led by Andy Acrivos,

among others, struck back. Their efforts led to the formation of the Fundamentals Section of the Program Committee, and a policy of 15-minute presentations with 5 minutes for discussion. This policy produced a revolution in fluid mechanics. I believe it is fair to say that as a result of this new policy the primary frontier in that subject shifted from all fields of engineering in all countries to the Annual AIChE meeting. Unfortunately, the change in format was far less successful in heat transfer as I will subsequently explain.

External Influences

Sources of investment in and support for advances in transport The political, social, and business climate in the USA has had a major impact on research and development in transport over the past century. Before the founding of the AIChE, the study of transport was most highly developed in Europe and its academic institutions. The early studies of transport in the USA were carried out in industry and often in the context of the operation of process equipment rather than research. Prior to World War II, most advances were prompted by new chemical processes and were discovered by the analysis of full-scale or semi-scale operations as well as by laboratory-scale experiments. During the war the same pattern occurred in petroleum refining. Since that time, both industrial research and academic research, the major contributors to advances in transport, have gone through several ups and downs.

At the end of the war the military services, recognizing the major impact that basic research had had on weaponry and other materiel, and awash in appropriations that had not yet been scaled back to peace-time levels, decided to utilize the excess funds for research rather than give them up. I was a beneficiary of such support for nearly a quarter of a century, and found it more enlightened and free of restrictions than any since. Those golden days came to an end in 1973 by virtue of the "Mansfield Amendment", which, provoked by opposition to the Viet Nam War, forbid *DOD* to support nonmilitary (non-mission-oriented) research.

Industrial research laboratories, such as Shell Development, Esso Research and Engineering, and Bell Laboratories, became pre-eminent in the post World War II years and attracted some of the best doctoral graduates in chemical engineering. They made significant fundamental contributions to transport, of which I will mention one that may not be known to all of you, namely the invention of zone refining in 1950, an essential step in the development of transistors, by Bill Pfann, a chemical engineer, at Bell Labs. Beginning about 1960, basic research fell out of favor with the corporate culture and these industrial research centers gradually lost their prominence and ceased their innovations, at least of a fundamental nature.

In the post World War II years, the chemical industry, and particularly the petroleum industry, was a major source of support for graduate work in chemical engineering in the form of fellowships. This unique source of support, which elevated graduate work in chemical above that in all other branches of engineering, was phased out beginning in the 1960s. The proffered reason for this withdrawal was the fulfillment of that need by *NSF* and *NASA*, but the real reason was their own withdrawal from research.

The direct support of academic research itself in chemical engineering by industry has had its ups and downs and is currently negligible. The first contract research was apparently carried out at the University of Michigan in the 1930s by Walter L. Badger for the Swenson Evaporator Company. Such industrially contracted research grew for a few years but soon faded because of competition from governmental research and proprietary concerns.

A significant event was the founding of the Heat Transfer Research Institute in 1962. Up until that time, the major chemical and petroleum companies sponsored a significant

amount of academic research in heat transfer. That sponsorship was promptly terminated along with their own research in that field. Most of the work undertaken by *HTRI* was quite applied, but because it was proprietary, an academic researcher in heat transfer, could no longer know whether or not the work he was contemplating was already completed or in progress. The impact was soon evident. Up to that point, the participation of chemical engineers in the US National and International Heat Transfer Conferences was roughly equal to that of mechanical engineers. Thereafter, it faded to nearly zero. Research in heat transfer by mechanical engineers survived because their work was supported by industries, such as those involved in aircraft and spacecraft, that did not join *HTRI*. One noteworthy exception was the contract research sponsored by Wolverine Tube, at the University of Michigan, which extended about 35 years under the consecutive direction of Alan Foust, Don Katz, and Ed Young, but that project too is now history.

The National Science Foundation came into being in 1950 but was not a major factor in academic research in chemical engineering until 1957, when *Sputnik* prompted panic over the state of research in the USA. *NSF* gradually became the major and essentially only source of funding for research in transport. Then in 1980 that source of support suffered a devastating blow, namely, the directive that all research sponsored by *NSF* should have a mission aligned with those of the “government” such as that of the *Strategic Defense Initiative*. If enforced rigorously, that directive would have had the effect of replacing the judgment of individual researchers as to what has promise with that of administrators. Even if the administrators of contracts and grants were “the brightest and best”, I would still have more faith in the productivity of exploratory research by individual researchers on the topics that they consider to be most promising. The impact of this policy has been aggravated by the lumping of funds into a few large grants on favored topics. The academic community is necessarily adept at coping with such policies, but innovation suffers. Do you believe that it is currently possible to obtain support for research on some aspect of transport without hiding it in work on energy conversion, the reduction of pollutants, nanotechnology, or biotechnology? Most of the cited advances in transport over the last 20 years have been from unsupported or “bootlegged” research.

Summary

I have presented a few illustrations of our progress over the past century in predicting transport. They are just that – illustrations; I have been highly arbitrary in my choices. Some of that progress consists of the abandonment of familiar concepts – a painful process and one that risks the appearance of criticism of idols of mine as well as yours. I trust that if they were here they would approve. In that regard, I recall W.K. Lewis, in an anecdotal lecture at an *AIChE* meeting, mentioning that the *Lewis number* commemorated his worst conceptual error.

Most of the advances that I have described today originated in academic research and most of the remaining ones in industrial research. However, I was able to identify them only because they have achieved sufficient recognition, acclaim, and certification to appear in textbooks and handbooks. The ultimate certification is their adoption for design, operation, and analysis, but that is difficult to quantify, except perhaps their “appearance” in computational packages. A few advances were mentioned that have a more limited but important role, namely improvement in understanding, and a few that had an even more limited objective, namely resolving some technical or scientific uncertainty.

Conclusions

Over the First Century of the *AIChE*, the most obvious improvement in the prediction of transport is a consequence of the development of powerful computer hardware and user-

friendly software. However, that advance in itself would have had little impact without three others

- 1) the formulation of theoretically based models for transport, primarily by non-chemical engineers
- 2) the formulation of special-purpose algorithms, primarily by chemical engineers
- 3) the interpretation of the computed values in theoretical and algebraic terms, almost wholly by chemical engineers

Two implicit elements should also not be forgotten

- 1) The accuracy of numerical solutions depends critically upon the validity of the model and on the convergence and stability of the methodology used for its solution.
- 2) Trusting models and/or their solutions whose limits of accuracy and validity have not been tested with experimental data is equivalent to believing in the Easter bunny.

The only reliable expressions for the prediction of transport are those that have a theoretical structure **and** have been confirmed by both experimental data and numerical simulations. Some of you who work in process design or operation may dismiss what I have said as mathematically oriented and once removed from practicality. That is a dangerous conclusion. In the early days of the AIChE correlating equations were devised by drawing a straight line through a log-log plot of experimental data. Over the century we have come to realize that expressions so-derived are almost certainly in error functionally, and thereby, outside a narrow range, may be in serious error numerically as well. If you are clinging to any such expressions involving products of arbitrary power-functions, for example the Colburn analogy, you are dangerously out of date.

The principal improvement needed with respect to the prediction of transport in conventional systems is the development of a methodology for the accurate prediction of developing turbulent flow and of convection in developing turbulent flow. The κ - ε model purports to fill this need but it is highly unreliable near the wall, which is the most critical region.

The current lack of interest in and support for research in transport is clearly a consequence in of the expansion of biological processing, a related expansion of interest in product as opposed to process design, and an extension of the scale of processing from the macro to the nano.

Bulk processing of chemical materials will undoubtedly remain an important part of our economy and energy conversion may spur a renewed interest in transport on the macroscale, but I suggest that we take advantage of the opportunity to expand the scope of transport to encompass biotechnology and nanotechnology.

We can take justifiable pride in the advances in transport during the *First Century of the AIChE*. I anticipate that comparable advances that we cannot yet imagine will occur in the *Second Century*.