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Wider horizons for fluid mechanics

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Preface

Readers of this Journal do not, in my experience, have a uniform view of what properly constitutes the discipline of Fluid Mechanics; nor would they agree on what the main outstanding problems in the subject are. Many, particularly among those who do not contribute to the Journal, believe – unfortunately – that it has become a branch of applied analysis on the theoretical side, and is dominated by hot-wire and laser anemometry on the experimental side. As an ex-editor, I must hasten to add that this has not been the view of the editors, and that, if such an impression is created by the spectrum of papers published in the Journal, such an imbalance largely reflects the nature of the papers submitted.

My thesis here is that it would be unwise to take too narrow a view of fluid mechanics, and that the directions in which useful progress might most easily, and most probably will, be made, lie along paths unfamiliar to most workers in the subject. The implication of this thesis is that the teaching of fluid mechanics in many engineering, applied mathematics and mechanics departments should be reassessed: the syllabuses chosen and the approach used should be examined critically in the light of new developments in the subject and of those situations, involving fluid flow, that most perplex those working primarily in other fields.

The teaching of formal fluid mechanics is now dominated by the continuum mechanics of a homogeneous viscous fluid obeying the celebrated Navier–Stokes equation and the continuity condition. Their applicability to a wide range of important gas and liquid flows is well established; analytical or numerical solutions based on them have been very successful in providing predictions for flow and pressure fields which can be verified experimentally. The attractions of this approach are obvious: (*a*) a minimum of basic assumptions and variables of interest; (*b*) a clear connection with physical reality on a visually obvious and familiar scale; (*c*) a clear and unambiguous mathematical formulation; (*d*) a graded set of formally rigorous mathematical solutions to idealized problems incorporating interesting nonlinear effects; (*e*) a well-documented record of past successes in anticipating (and not merely explaining after the event) important practical observations.

One well-known weakness of this approach lies in the field of turbulent flow. Despite remarkable successes in predicting instability of laminar flow solutions and in investigating regimes where multiple solutions exist, formal exact theory cannot treat fully the turbulent flow which characterizes many situations of interest in engineering. Such treatments as are now used involve a series of additional empirical or phenomenological assumptions, which lead in most cases to complex numerical solution of engineering or geophysical flow problems. It is noticeable how few of the contributors to this volume have chosen to write about turbulence, despite early endeavours in that field by many of them. It is no exaggeration to say that a rational theory of turbulence remains the outstanding challenge in fluid mechanics.

Strivings towards a formal theory of turbulence based directly on the Navier–Stokes equation did, however, lead to incorporation into classical fluid mechanics of many important statistical ideas, which are shared with statistical mechanics. For example, distinctions between ensemble, time and space averages, though no longer widely discussed in fluid mechanics, remain lively matters of interest in the theory of liquids, and have relevance in current environmental problems of effluent or pollutant dispersion. The subtle role of 'internal' length and velocity scales, and the spectral (eddy) approach, are further concepts that extended traditional approaches to fluid mechanics, and are of importance in rheologically complex fluids. It is a pity, therefore, that formal teaching of fluid mechanics does not emphasize these aspects more and so direct students towards the underlying ideas that those who have worked on these problems regard as essential.

Concentration on laminar flow solutions for homogeneous Newtonian fluids has other disadvantages. It obscures the central role implicitly or explicitly played in a fluid-mechanical theory by the constitutive equation (rheological equation of state) relating the local stress tensor to the history of deformation (as well as certain other variables). It tends to separate the mechanics of flow from the thermodynamics, instead of stressing the essential unity and breadth of a full continuum theory. This separation of approach is abetted by many teachers of thermodynamics, who concentrate on equilibrium states, and regard transport phenomena as disjoint from their field of interest.

The following sections are written from the point of view of a chemical engineer, for whom fluid mechanics plays a less central role than for an aeronautical or civil engineer, though not, in my view, a less important one. It is the connections between fluid motions and other aspects of engineering that are of most importance in applying formal theories and basic results on fluid mechanics.

Fluid mechanics in chemical engineering

Fluid mechanics is taught in all chemical engineering courses. It presents more difficulty in my experience than any other subject in the curriculum (Pearson 1980). It is widely regarded as an essential element in engineering practice, although I have come across relatively few practising chemical engineers who make creative use of fluid mechanics (and, for many of them, this ends with the Navier–Stokes equation) in solving engineering problems. In most cases, they make cautious use of the many engineering correlations that have been enshrined in the most widely accepted textbooks, such as for friction factor in pipe flow, or terminal fall velocity of freely falling particles; at worst they disregard all fluid-mechanical effects as such, incorporating them implicitly into heat- or mass-transfer correlations. Because of this, they fail to gain insight into many processes of interest and importance, and fail to anticipate or solve problems that arise in their processes. They are not to be blamed for this, for their problems are complex and the issues most important to them are rarely considered by experts in fluid mechanics in a context familiar to them.

Mixing in liquids – blending, agitation, dissolution, emulsification, or just plain stirring – is a typical such issue. The process is inseparable from the mechanics of the flow employed; in many cases, the rheological properties of the fluid (relevant in the momentum equation) are dependent on the local proportions and state of the various constituents of the mixture, i.e. on the degree and uniformity of the mixing. So there can be strong coupling between the mixing flow and the material being mixed. This is well known in a general sense to engineers, but there is little formal theory for them to refer to. Danckwerts (1952) provided a lead by proposing a statistical description of mixedness based on ideas then current in turbulence theory, distinguishing between scale and intensity, while Batchelor (1959) and Batchelor, Howells & Townsend (1959) investigated the passive mixing effect of turbulent motions. There, with a few honourable exceptions largely in the field of combustion (see reviews of Hill 1976 and Libby & Williams 1976), the subject has rested, or so it would appear from the chapters on mixing in most engineering texts. Most attention is still paid to power factors and algebraic scaling laws, it being implied that industrial needs can be met by selection from a small range of agitators – paddle, turbine, helix, propellor – and baffled vessels, based on elementary considerations and past experience. In practice, however, a great many other devices are used to produce mixtures, but these are usually described by special terms – atomizer, homogenizer, plasticizer, mill, carburettor – each with their own folklore, and are not recognized primarily as mixers. Seldom is a student presented with much overall insight into the differing contributions made by different flow fields – imposed laminar, imposed turbulent and naturally unstable – or into the objectives of the mixing processes, which may include heat transfer and aggregation or disaggregation of a dispersed phase.

A difficulty I have met when co-operating with many practising engineers is in persuading them to go far enough back into the processing problem in question; all too often they assume, without discussion, that they have correctly formulated its fluid-mechanical element; and inexperienced workers anxious to display their analytical or experimental skills in fluid mechanics happily embark on extensive investigations, convinced that their work is relevant without sufficient appreciation of the process as a whole. The proper course,† less often followed with care, is (*a*) to define as unambiguously as possible, in each case, those variables that are important in the product (and this often involves a still different group of people), and to state the values or range of values that they should have in an acceptable product; (*b*) to define those input and operating variables that characterize the process and which may be adjusted to achieve the desired product; (*c*) to provide a physical and, where possible, a mathematical model relating input to output variables.

It is at this stage that the fluid mechanics governing the process becomes important: any model will involve a knowledge of the velocity, temperature, and – in some cases – stress, fields characterizing the process. Skill is, however, required in deciding which parts and which aspects of the flow field are important in the coupled set of governing equations, and the level of accuracy required in describing the relevant portions of the flow field; in recognizing whether steady-state or transient effects are the primary cause of difficulty. For example, many difficulties only arise at the entry or exit

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† It is not that this is not recognized by many teachers of engineering fluid mechanics, and indeed open-ended problems set are often selected to illustrate and develop this approach. Unfortunately the view that students should have some knowledge of a vast range of scientific theory and should be able to reproduce a lot of standard techniques allows little time in most curricula, which are based on clearly identified subject headings, to achieve the necessary depth or breadth of approach in tackling relatively complex industrial processes.

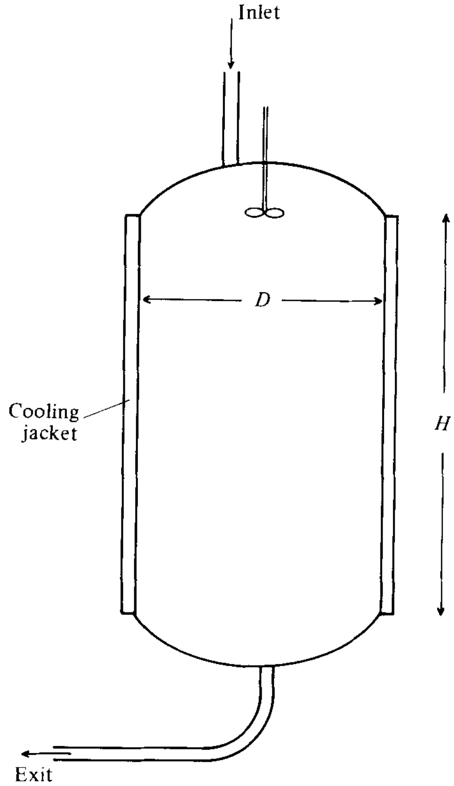

FIGURE 1. Diagram of continuum flow PVC reactor that is kept full of liquid.

regions: elaborate bulk analyses that idealize these regions by prescribed inlet and exit boundary conditions will never provide significant understanding.

Unfortunately, examples of the successful application of fluid mechanics to engineering problems do not conveniently lend themselves to publication as original papers. Such work as does appear in print is often suggested by the original problem, but is needlessly elaborate for practical purposes; indeed, one can go further and argue that its nature and depth is more determined by national arrangements for supporting research students than any intrinsic need for, or significance of, the results published On countless occasions, as a referee, I have read introductory paragraphs which expound generally on the importance of work on, say, suspension mechanics to various industries (coal, paper, oil exploration, printing, paint), without any particular situation or problem being mentioned. What follows then makes little or no contribution to either the fundamentals of fluid mechanics or its application to a live problem involving other features.

An illustrative example (that is unlikely to reach the literature in any other context), of how a relatively broad knowledge of fluid mechanics can be usefully applied to explain an apparently paradoxical observation in reactor engineering, is appropriate. It concerns the continuous polymerization of a concentrated emulsion of vinyl chloride in an aqueous solution to yield a fine suspension of PVC particles. A very simple schematic diagram of the reactor vessel claimed in a German patent to achieve the desired result is shown in figure 1. The interesting feature is that only a small stirrer is shown at the top (inlet) of the cylindrical vessel of large (many metres) diameter and even greater height. The polymerization process requires

(*a*) that the time spent in the reactor by each element of monomer/polymer depart as little as possible from a known mean, i.e. the reactor should behave as a 'plug-flow' reactor with a sharp residence-time distribution;

(*b*) that the temperature throughout the reactor be kept as close as possible to a fixed operating temperature, i.e. within a few degrees of some value between 50 and 75 °C;

(*c*) that there be no segregation of the phases.

Important features of the process are:

(*d*) that a large amount of heat is released by the polymerization reaction, and so strong cooling has to be applied;

(*e*) that polymer is significantly denser than monomer, and so the mean density of the emulsion/suspension in the reactor increases from top to bottom if plug flow is achieved;

(*f*) that a stable emulsion is achieved in the input by surface-active agents and that polymer accumulates around sites initiated in very small micelles of vinyl chloride, i.e. polymer particles form separately from the main droplets of vinyl chloride, and the monomer diffuses through the aqueous phase from droplet to growing particle.

The arrangement, if successful, presents a paradox to an engineer asked to assess the merit, or even the feasibility, of the design. How can plug flow be achieved, i.e. flow characterized by the absence of vertical mixing, together with efficient radial heat transfer? Molecular transport of heat can be discounted as relevant except in very narrow boundary layers (of order 1 mm thick) at the surface of the reactor, while any imposed radial stirring is ruled out on mechanical grounds by the size of the reactor. Indeed the diagram shown includes but a single small stirrer at the top used to ensure an initially satisfactory emulsion. Those familiar with natural thermal convection will recognize at once the possibility of large convection cells arising as radial temperature gradients tend to develop. However, a convective pattern such as the one shown in figure 2(*a*) would tend to make the reactor into a single well-stirred one with an exponential form of residence time distribution and therefore unacceptable according to (*a*) above. At the same time, the mean density gradients due to polymerization, mentioned in (*e*) above, will tend to stabilize the system against vertical motions. Those familiar with geophysical applications of fluid mechanics will readily recognize that the multicellular structure shown in figure 2(*b*) would then be the most likely. The paradox is finally resolved by recognizing that the four (or more) cells shown could be crudely regarded as fully stirred reactors in series; which reactor theory has independently shown to be little different in practice from a true plug-flow reactor.

Note that so far only general ideas relating to flow patterns have been put forward. Once a likely flow pattern has been proposed, i.e. as in figure 2(*b*), crude calculations can be carried out to estimate flow velocities and heat transfer rates. The details of even an order-of-magnitude approach go rather beyond traditional textbook treatments. In the case illustrated, it was possible to show that natural convection could account (at a Grashof number of unity) for the necessary heat transfer. There remained the question of estimating the number of cells likely to develop. A systematic search among recent *Journal of Fluid Mechanics* titles discovered several papers which

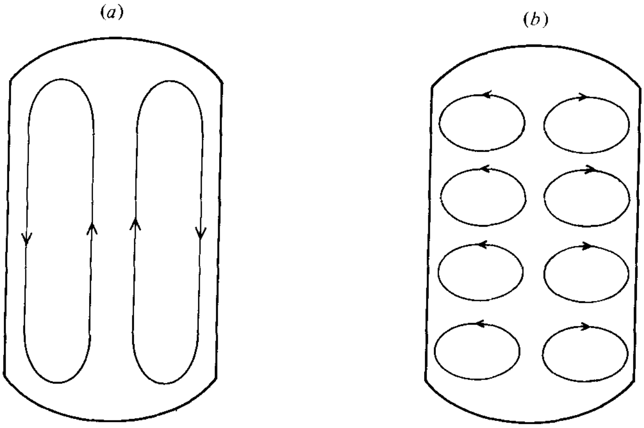

FIGURE 2. Convective patterns driven by density gradients: (a) unacceptable as would be expected with no irreversible material density changes; (b) acceptable, due to mixed thermal and irreversible material density gradients.

addressed related situations, in which density gradients were present, and presented experimental data. This provided strong support for the multicellular model.

The engineering analysis briefly described above was carried out in a relatively short time to test the feasibility of a design, which departed remarkably from traditional batch process designs, and which achieved an increase in scale only by an apparent disregard of standard scaling laws, particularly as regards agitation for heat transfer. Any attempt to simulate the reactor flow on the basis of a complete numerical treatment of the coupled equations would have taken far longer and been too expensive; direct laboratory-scale experiments would have yielded nothing of relevance in the time available. Even if the feasibility study were to be pursued to provide more precise quantitative information, it is highly probable that both theoretical and experimental work would be based on a multicellular model. The issue that is relevant to my thesis is whether traditional teaching of engineering fluid mechanics would lead even relatively gifted engineers to carry out such an analysis, or to know how to present information to more specialist workers in fluid mechanics in such a way as to lead them to appreciate the critical aspects of the problem. My experience, not just in this particular case, but over a wide range of complex problems, is that it does not.

Rheological equations of state

Industrial processes, particularly those considered by chemical engineers, have provided many examples of the flow of fluids that are clearly non-Newtonian. Polymeric systems have been the most widely studied class, but dense suspensions of particles or drops are equally important and interesting. More general mixtures, reacting systems and those showing phase changes further extend the range. The importance of describing such materials rheologically has been repeatedly stressed for the last four decades and major advances have been made in continuum-mechanical theories. The pioneering paper of Oldroyd (1950) deserves separate mention. Different aspects of the subject are more than adequately treated in various texts: an exhaustive general account is given in Truesdell & Noll (1965); a class of relations relevant for rubber-like fluids is given in Lodge (1964), while a more comprehensive treatment for polymeric liquids is given in Bird, Armstrong & Hassager (1977); a much briefer account is given in Astarita & Marrucci (1974). The last three concentrate wholly on the relevant constitutive equations for a homogeneous isotropic continuum, while the first deals largely with homogeneous materials. Boundary or jump conditions are rarely discussed in any detail.

It is quite clear that all real materials of interest show profound inhomogeneity, and considerable structure, when viewed on a small enough scale. It is a basic problem of physics to explain observed behaviour in terms of more fundamental physical laws applied to the motion of the structural elements. If the element is taken to be a molecule (now regarded as the realm of chemistry), then the techniques of classical statistical mechanics are employed. However, as Batchelor (1967, p. 53) has written, even for very simple molecular fluids, "much less is known about the structure of liquids than about that of gases. No simple model, like that of a perfect gas with dynamically independent molecules, is available for the derivation of approximate results for liquids. As a consequence, it is not possible to set many of the observed values of properties of a liquid within a logical framework or to account for them in terms of properties of the individual molecules". In particular, the linear (low shear rate) viscosity is thus treated as a parameter best evaluated by experiment. The only simple molecular theory of liquid viscosity is that provided by Eyring (Eyring 1936) and developed with coworkers based on the jumping of molecules over energy barriers between equilibrium positions which leads to a non-Newtonian viscosity which behaves as $(\ln D)/D$ for large shear rates D, although it can be regarded as constant for observable shear rates. Attempts have been made to employ more rigorous molecular dynamical calculations to the prediction of a rheological equation of state: difficulties, that are largely concealed rather than resolved in the Eyring treatment, arise in connection with the definition of the stress tensor in a non-equilibrium configuration, most particularly near interfaces, where similar difficulties arise in the definition of equilibrium thermodynamic functions. In practice, of course, simple fluids are well described by an easily measured viscosity.

More progress has been made in the case of some more complex fluids. One class is provided by dilute suspensions of solid particles, which are modelled in terms of a continuous linear viscous phase and a rigid (or visco-elastic) solid phase with continuity of velocity and stress across the interfaces; a very similar class is provided by dilute emulsions consisting of a dispersed viscous (or elasticoviscous) liquid in a continuous linear viscous phase separated by interfaces exhibiting constant surface tension. The size of the particles or droplets is large compared with molecular dimensions. Here the technique has been to study the exact flow around the particles analytically by means of Newtonian fluid mechanics and then to use statistical (averaging) arguments (Herczyński & Pieńkowska 1980) to obtain the continuum equation of state for the suspension on a scale large compared with the separation distance of the particles. Results have been obtained for non-spherical particles, two-particle interactions and Brownian motion (rotational diffusion) of particles. Batchelor (1976) provides a review of these ideas, and notes that the derivations and the equations of state are as exact as any calculations made in traditional fluid mechanics for Stokes flow of a Newtonian fluid in a region bounded by solid or deformable surfaces. Many of the logical steps employed are analogous to those used in molecular dynamics and statistical mechanics. Instead of Newtonian particle mechanics allied to intermolecular potentials which are used in molecular dynamics, continuum mechanics is used in synthetic rheological theories for suspensions. Unfortunately, the methods employed cannot yet be extended rigorously to include dense suspensions, though very interesting proposals have been made (Goddard 1977) that involve lubrication theory and a denumerable set of effective point contacts between particles.

The other major class of complex fluids that has been successfully investigated is provided by polymeric fluids, which are conveniently discussed under three categories: dilute solutions, concentrated solutions and homogeneous polymer melts. The polymer molecules are interesting because they exhibit two widely different length scales: that of the monomer unit in the chain which is of molecular dimensions similar to those considered in molecular dynamics, and that of the extended chain, which is many orders of magnitude larger, though still small compared with the fluid-mechanical length scale. For entropic reasons, flexible polymer molecules assume a coiled form, with a characteristic diameter that is the geometric mean of the monomer and extended chain lengths.

The dilute polymer solution is treated in some ways like a dilute suspension. Because of the connectivity requirement along the chain, most parts of the chain must be moving relative to the solvent in any sheared flow, the latter being assumed to have the local mean fluid velocity (the 'affine' assumption). The mechanical interactions between polymer chains and surrounding solvent are then modelled by considering a pseudo-chain of beads and springs (or rods); the springs (or rods) move freely in the solvent while the beads suffer a Stokes drag force. The model chain is defined by the number of beads, the strength of the springs and the drag coefficient of the beads; Brownian forces can be included. Such theories have recently been fully described in Bird, Hassager, Armstrong & Curtiss (1977, chapters 10–14) who stress the role of kinetic theory in obtaining rheological equations of state. These are of the elastic-viscous type introduced by Oldroyd (1950). The details are relatively complex, and lead to a range of predictions for the stresses in controllable† flow fields.

A concentrated polymer solution is one in which the polymer molecules overlap and interact directly. Models for these are more varied. However, the most common are continuous network models, in which the key notion is that of a 'junction', i.e. a point at which there is a temporary connection between two chains. Between junctions the chains are modelled by springs, with the same properties as in the bead–spring model. If the junctions were permanent, and all interconnected, the material would be a swollen solid rubber. If the concentrated solution is to possess fluidity, the instantaneous network must be capable of breaking and reforming. The simplest account is given in Lodge (1964), who adopts the 'affine' concept for the movement of junctions; the basic model and various extensions are described in Bird, Hassager, Armstrong & Curtiss (1977, chapter 15), although several more have since been proposed.

Molten polymers are the most important class industrially, but they have been less carefully investigated in terms of a full rheological equation of state. A recent theory of Doi & Edwards (1978) based on a reptating chain-in-tube model has stimulated a

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† Controllable flows are those in which a known flow field can be imposed by the deliberate movement of rigid boundaries whatever the rheological properties of the fluid; it is assumed that inertial forces are negligible, but controllability also assumes that the flow obtained will display the maximum degree of symmetry consistent with the boundary conditions; unfortunately such flows can become unstable, with less symmetric, but often steady, flows then taking their place.

re-examination of this problem and careful comparison of the rheological equations of state provided by various apparently independent theories. In particular, Curtiss & Bird (1980) have shown that a bead–rod model leads to essentially the same equation of state as that of Doi & Edwards; furthermore, it is of the same general type as those given by temporary network theories.

A recent text by de Gennes (1979) provides very considerable insight into polymeric fluid behaviour generally. In particular, the importance of the blob length scale is made clear: within a blob, the effect of excluded volume is important and so an individual chain interferes with itself; over lengths larger than a blob diameter, a chain can be considered a Gaussian (phantom) chain of blobs. The blob thus introduces a length scale larger than the monomer and smaller than the diameter of the entire chain. Thus the physics of a coiled chain introduces of itself a length that removes some of the arbitrariness associated with chains of beads (i.e. their diameter and mean separation), with tubes surrounding reptating chains (i.e. tube diameter) and with networks (i.e. distance between junctions).

Batchelor, Bird, Edwards, de Gennes and their many co-workers have done a singular service to fluid mechanics, by emphasizing its connections with statistical mechanics of the fluid state, and by presenting theories for structured fluids (including liquid crystals) in a form which incidentally satisfies the invariance requirements of formal continuum theories. They have taken rheology beyond the phenomenological backwater of linear viscoelasticity into which many of its practitioners had retreated.

Boundary conditions

Continuum theories admit of jump discontinuities of properties and so solution of the field equations of continuum mechanics requires boundary conditions to be specified at the relevant interfaces. Apart from general agreement on the operational use of the concept of surface tension between two (immiscible) bulk liquids, there has been relatively little fruitful contact between fluid mechanics and surface physics and chemistry. In general this had led to little difficulty, because the no-slip boundary condition at solid surfaces together with continuity of stress and velocity at free surfaces (special account being taken of the normal stress discontinuity associated with surface tension acting over a curved surface) have been adequate for most problems in fluid mechanics.

However, there are exceptional cases where difficulties arise, and where a great deal of confusion exists in the literature. Thin liquid films, as in foams or on surfaces, can exhibit apparently non-Newtonian properties that are not characteristic of the same liquid in bulk. Concentrated emulsions containing dissolved polymers or polar surface-active agents can similarly display behaviour uncharacteristic of either bulk phase or of simpler emulsions of the same concentration. Theoretical analyses of flow fields involving sharp corners or moving contact lines (lines at which the interface between two immiscible fluids meet a solid surface) according to traditional Newtonian fluid mechanics often predict singularities in stress; these are in practice not observed.

Molecular dynamical investigations have shown that the simplest plane liquid/vapour or liquid/liquid interfaces are in fact very thin layers of order 5 molecular diameters thick within which large difference-of-normal stresses arise. This is illustrated in figure 3. Within the bulk of fluid A, the stress at equilibrium $\mathbf{T}_A = -p_A\,\mathbf{I}$ is

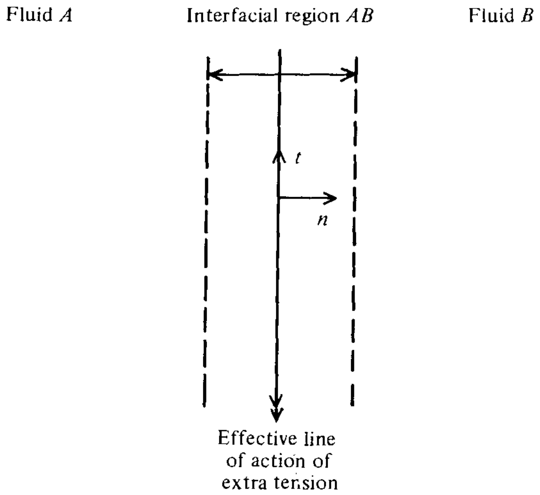

FIGURE 3. The Gibbs representation for a fluid/fluid interface.

isotropic; within the bulk of fluid B, similarly, $\mathbf{T}_B = -p_B \mathbf{I}$; within the interface region, the stress $\mathbf{T}_{AB}$ can be written, on the basis of planar symmetry, as

$$\mathbf{T}_{AB} = -\begin{bmatrix} p_{nn} & 0 & 0 \\ 0 & p_{tt} & 0 \\ 0 & 0 & p_{tt} \end{bmatrix},$$

where $p_{nn} > p_{tt}$ for immiscible liquids; $\mathbf{T}_{AB}$ is a function of n and not t. Gross equilibrium requires that $p_A = p_B = p$, it being assumed that intermolecular forces (both long and short range) act effectively only over distances of the order of the layer thickness. The surface tension is defined by

$$\gamma_{AB} = \int_{-\infty}^{\infty} dn(p - p_{tt}).$$

If we seek to calculate p_{nn} and p_{tt}, which are continuum quantities, it becomes clear that the overlap of the molecular and what is now the continuum scale for the region AB leads to difficulties. A halfway-house approach is to neglect the actual motion of molecules, replacing their thermal energy everywhere by a uniform isotropic pressure $\frac{1}{2}k_B T$; the equilibrium situation then becomes a static one whose stress field is determined by means of a continuous representation for the pairwise intermolecular potential field. The real situation involves sharp mean-concentration gradients in the inter-facial region, with concomitant thermodynamical driving forces; the equilibrium concentration distribution would in thermodynamical terms be determined by the requirement of constant chemical potential. However, it would seem that the calculation of all thermodynamical variables in such an interfacial region is subject to the same uncertainties as that for the stress. A more tractable situation is apparently provided by the case of almost miscible liquids, i.e. an interfacial region of very low excess surface energy (surface tension), when the interfacial region becomes arbitrarily thick in terms of the molecular length scale, though formally still arbitrarily thin on the continuum length scale.

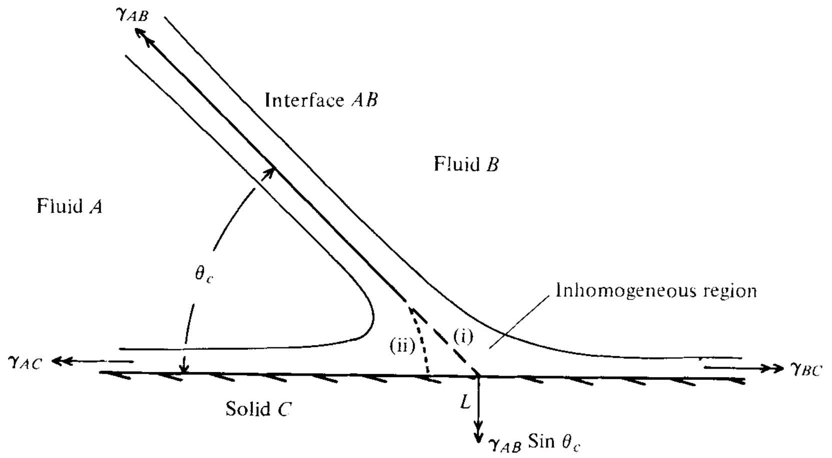

FIGURE 4. The static contact line.

The next stage is to consider the case of an equilibrium contact line† as shown in figure 4. The continuum representation is to suppose that the plane interface AB meets the solid surface at the contact line L, subtending an equilibrium contact angle θ_C; Young's equation states that a balance of forces in the plane of the solid surface provides a relationship between γ_{AB} and the two surface excess energies γ_{AC} and γ_{BC}, namely

$$\gamma_{AB} \cos \theta_C = \gamma_{BC} - \gamma_{AC}.$$

Note that γ_{BC} and γ_{AC} can be defined in terms of T_{BC} and T_{AC} just as γ_{AB} was defined in terms of T_{AB}. If Young's equation is correct, it requires that a normal force/unit length $\gamma_{AB} \sin \theta_C$ act on the solid C at L. No widely accepted explanation (see Dussan V. 1979, pp. 377–380) has been given as to why this force should be normal, and contact-angle hysteresis after motion of the contact line suggests that in practice it will not always be so. Resolution of this matter in terms of a local continuum treatment for the non-homogeneous (in terms of the concentrations of A, B and C and out-of-balance molecular forces) region near L has led to differing predictions. In particular, no unique means of definition of the interface AB in the neighbourhood of L has been accepted. The broken line (i) shown in figure 4 is simply an extrapolation of the Gibbs interface into the region where T_{AB} will vary with t as well as n (t and n as defined in figure 3); there is no reason to believe that the dotted line (ii), for example, might not be as realistic if a sharp interface were demanded by traditional continuum-mechanicists anxious to maintain the stark simplicity of field equations plus jump conditions.

Steady motion of the contact line, which has been observed experimentally, presents additional problems for the continuum-mechanical approach. If a planar interface AB is, as before, regarded as meeting the solid surface in a contact line L (figure 5) then any change in the contact angle $\theta_C^*(U)$, now regarded as a function of the speed U of the solid surface relative to the contact line, will upset the balance of forces shown in figure 4. For two strictly inviscid fluids at constant pressure, a force balance would require a contact line force γ_{LC} that was no longer normal to the

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† The account given here is described much more fully in the review of Dussan V. (1979), which gives a most useful list of references.

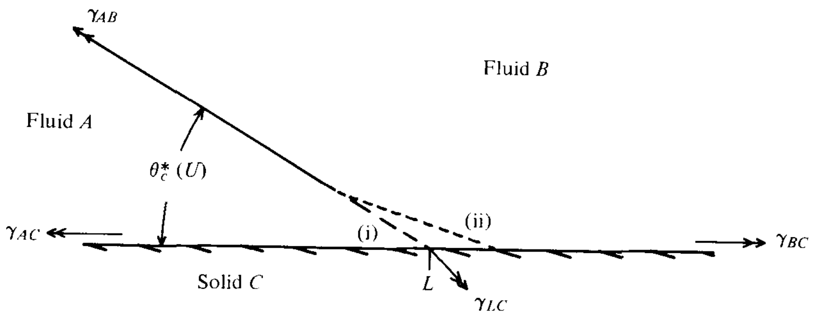

FIGURE 5. Moving contact line (solid going with speed U from left to right with respect to L).

surface of C if $\theta^*(0) \neq \theta^*(U)$. The additional components along the surface (assuming Young's equation to hold) could be regarded as a velocity-dependent frictional force

$$\gamma_{LF} = \gamma_{AB} \cos\{\theta_C^*(U) - \theta_C^*(0)\}$$

associated locally with the contact line. For highly viscous Newtonian fluids, the situation is much more complicated because the viscous stresses necessarily created by the velocity fields $\mathbf{v}_A$ and $\mathbf{v}_B$ would lead to deformation of the interface AB. Furthermore any velocity and stress fields satisfying the continuity equation and a no-slip boundary condition at the interfaces AB, AC and BC lead to singular stresses at the contact line (Dussan V. 1979, pp. 390–393), which destroy any simple interpretation of the local contact angle. Thus in figure 5, the interface AB would have infinite curvature at L, and could not be treated as straight near L.

Two remarks, which go beyond those made in Dussan V. (1979), should be made at this point. The first is that the only absolute theory of liquid viscosity, that of Eyring mentioned earlier, does not display this singularity because the shear stress only rises logarithmically with shear rate; the second is that a wall-slip coefficient based on the Eyring rate theory with comparable activation energies would allow of significant slip in the inhomogeneous region near L, and hence eliminate velocity discontinuities, even though the no-slip boundary condition would be accurately applicable far from L. It is thus perfectly reasonable to suppose that the continuum hypothesis could be retained even when providing a mechanical model for the moving contact line, provided a non-Newtonian viscosity and a small slip coefficient were used.

It will be readily recognized that the contact-line problem discussed above is idealized. When molecular dimensions are involved, real surfaces are not plane, nor are they clean, nor are they always uniform. It is by no means certain that contact line movements are steady. When non-equilibrium effects arise over such short length scales, the applicability of linearized irreversible thermodynamics (as expressed in terms of transport coefficients added to equilibrium state equations) is in doubt. However, the added complications introduced by these real effects should not distract attention from the logical and conceptual requirements of a continuum theory. If such a theory exhibits inconsistencies, these should be eliminated by the introduction of the least number of additional features (Occam's razor). A suitable marriage of physical and fluid-dynamical points of view has not yet been arranged, let alone solemnized.

Flow fields for complex fluids: a challenge to analysts

Much of the impetus for studying the rheology of polymeric and other complex fluids came from observed departures from Newtonian flow, such departures often being spectacular and puzzling (see, for example, Bird, Armstrong & Hassager 1977, chapter 3, or Lodge 1964, chapter 10); these include a strong tendency for elastic fluids to climb up a rod rotating in a bath of fluid, to swell by an order of magnitude when extruded into air through a narrow tube, to be capable of passing continuously from an open reservoir to a syphon tube with its upper end held above the free surface, to recoil when an extending filament is cut, and to display large regular or irregular oscillations in flow when extruded from a reservoir through a narrow tube. In all cases, these effects grow with increase in flow rate. If the fluid can be described by a characteristic time λ and the flow field by a characteristic time τ (whose inverse might be the deformation rate D or the relative rate of change of deformation rate $\dot{D}/D$) then a dimensionless measure of the importance of elastic effects is given by the Deborah number

$$De = \lambda/\tau.$$

The effects mentioned usually arise when De becomes appreciable, and are most spectacular when De is large compared with unity.

Fluid mechanics is remarkable for the detailed analytical and numerical solutions obtained for laminar flow of Newtonian fluids in a wide range of geometries. The same cannot be said to hold for rheologically complex flows, except when departures from Newtonian behaviour are relatively small, and a perturbation about the Newtonian solution can be carried out. Examples of such solutions have been provided by Rivlin and co-workers (see, for example, Rivlin 1979) and by Joseph and co-workers (see Joseph & Beavers 1977, for a survey). The advantage of low-Deborah-number expansions is that the rheological equation of state can be universally expressed as a Coleman–Noll retarded-motion expansion, material properties being expressed through definite values of a set of scalar constants. High-Deborah-number flows have not been successfully treated analytically, except for the rather simple cases of fibre extension (see Petrie 1979, chapters 5 and 6, for a comprehensive account), radially symmetric (sink) flow, and one isolated solution for wedge flow (Hull 1981).

Many workers have taken the view that numerical methods offer the best means of predicting flow fields for elastic fluids (see Walters 1979, pp. 118–120). Early work has tended to concentrate on plane or axisymmetric flow into contractions and around spheres or cylinders; indeed, it has recently been proposed that certain test geometries be agreed upon for comparative purposes. Equally, relatively simple rheological equations of state have been used, mostly of the differential or rate type; for example, the Oldroyd 8-constant model

$$\begin{aligned}\mathbf{T}^E + \lambda_1 \frac{\mathscr{D}\mathbf{T}^E}{\mathscr{D}t} + \mu_0(\mathrm{tr}\,\mathbf{T}^E)\mathbf{D} - \mu_1(\mathbf{T}^E.\mathbf{D} + \mathbf{D}.\mathbf{T}^E) + \nu_1(\mathbf{T}^E{:}\mathbf{D})\mathbf{I}\\ = 2\eta_0\left[\mathbf{D} + \lambda_2\frac{\mathscr{D}\mathbf{D}}{\mathscr{D}t} - \mu_2\mathbf{D}^2 + \tfrac{1}{2}\nu_2(\mathbf{D}{:}\mathbf{D})\mathbf{I}\right],\end{aligned}$$

where $\mathbf{T}^E$ is the extra stress tensor and

$$\frac{\mathscr{D}}{\mathscr{D}t} = \frac{\partial}{\partial t} + (\mathbf{v}.\boldsymbol{\nabla}) + (\mathbf{W}.-.\mathbf{W}),$$

with

$$\mathbf{W} = \tfrac{1}{2}\{\nabla\mathbf{v} - (\nabla\mathbf{v})^{\mathrm{T}}\}, \quad D = \tfrac{1}{2}\{(\nabla\mathbf{v} + (\nabla\mathbf{v})^{\mathrm{T}}\},$$

is the corotational time derivative, has been widely used, with various values of the constants λ_1, λ_2, μ_0, μ_1, μ_2, ν_1, ν_2 and η_0 (the terms ν_1 and ν_2 effectively disappear when an additional arbitrary isotropic pressure $-p\mathbf{I}$ is added to yield the total stress tensor $\mathbf{T}$). Some particular cases of this model can equally well be expressed in integral form, e.g. the Oldroyd model B

$$\mathbf{T}^E = 2\epsilon_0 \frac{\lambda_2}{\lambda_1}\mathbf{D} + \frac{2\eta_0}{\lambda_1^2}\left(\frac{\lambda_2}{\lambda_1} - 1\right)\int_{-\infty}^{t} \gamma'_{[0]}\, e^{-(t-t')/\lambda_1}\, dt',$$

where $(\mathbf{I} - \boldsymbol{\gamma}_{[0]})$ is the Finger strain tensor. Experience so far has shown that iterative solutions do not converge satisfactorily for large values of the Deborah number. On physical grounds it can be argued that integral models should suffer less from numerical instabilities than differential models, but no conclusive evidence has yet been published on the underlying connections between Deborah number and numerical instability.

The challenge to workers in the field is twofold: first to select a single rheological equation of state that will adequately represent the behaviour of some important polymeric fluids over a wide range of unsteady deformation rates; secondly, to solve the resulting conservation equations for non-trivial flow geometries. It is to be expected that approximation schemes based on simple kinematical, dynamical and rheological models will be necessary to solve the relevant highly nonlinear sets of equations.

Flows involving large temperature (and/or pressure) gradients in fluids of low thermal conductivity and high viscosity also present unusual features when the rheological equation of state is highly temperature (and/or pressure) dependent. An interesting way of representing such rheological behaviour in polymers by rate-equations has recently been provided by Acierno *et al.* (1980). Many practical situations in polymer processing also involve phase changes (solid/liquid crystal/liquid) which may themselves be rate controlling; in the latter case, care has to be exercised in defining the relevant jump conditions at interfaces.

Other flows, of very considerable importance in chemical engineering, involve mass transfer by bulk diffusion and across interfaces. If the diffusing material is surface active, then variations in its concentration can strongly affect interfacial variables like surface tension. The mass transfer equations are then strongly and nonlinearly coupled to the flow equations. A currently important example of such a situation is provided by the two-phase flows in porous media characteristic of tertiary oil recovery.

Some readers may ask at this point whether there is any benefit to be gained by those steeped in traditional fluid mechanics tackling these physically more complex problems. I think that there is. Old-fashioned fluid mechanics is based on rigorous mathematics, on precise definitions, on careful analysis of error, on elegance in formulation, on a search for self-consistency and on comprehensiveness in approach within a chosen field. These characteristics are less evident in the wider field I have described; independent schools of thought flourish apparently tackling the same problems, but using disjoint empirical bases often incorrectly described as theories. Continuum mechanics is no longer a significant part of the education of physicists or chemists, and so their approach is often highly specific to their chosen materials or phenomena.

Successful practitioners of fluid mechanics are well schooled in the solution of non-linear problems, and have far more background in stability theory (or bifurcation theory) than all but a small proportion of engineers. Yet it is precisely the effects of instability, multiple solutions and hysteresis that are so important in natural and industrial processes. Those who have applied a strong fluid-mechanical background to complex problems have, in many cases, been outstandingly successful as evidenced by occasional fascinating papers in this Journal. The spirit of Maxwell, Stokes, Rayleigh and Taylor, to name but those most revered in this Journal's home base, will surely light upon equally distinguished successors.

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