1971 Award Lecture

THE ART AND SCIENCE OF RHEOLOGY*

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I. INTRODUCTION

When I began seriously to think about the content of this lecture, it occurred to me that perhaps the combination of an ASEE meeting, a depressed state of the engineering and science community, and an opportunity for me to speak on a subject of my own choosing all argued for a technical talk somewhat different from the format to which most of us are accustomed.

I do intend to talk about rheology, but I wish to use it as a vehicle for expressing some opinions on a larger subject; namely, the proper interaction, as seen by this writer, between technological art and engineering science. I hope that listeners will not consider such a topic to be inappropriate. Certainly part of the engineer's function is to achieve a proper match between the art and the science of his technical field. Rheology is an apt example for the matching and mismatching—of art and science since the subject spans axiomatic continuum mechanics and the processing problems of the plant polymer engineer.

I wish to cite examples of the ways in which the science and art of rheology need each other. The former brings order and understanding to the latter. The latter provides an essential motivation to the former. I shall have reached my goal if I persuade some that in spite of tight industrial research budgets and shrinking academic funds for non mission-oriented research, advanced research can be relevant-more so, in fact, than some development programs which proceed with no effort toward broadening our knowledge of the subject under study. At the same time I hope to convince others that new knowledge is not necessarily worth having simply because it is new. Indeed, we seem to be in danger of suffocating from an oversupply. I fear that some of the reaction that we see today against fundamental research is simply a consequence of the huge supply of engineering research which has appeared and proved neither fundamental (in the sense that it really brought us to a higher level of understanding of a subject) nor applied (in the sense that it led to a useful application).

I shall return to this point later but wish now to proceed to more technical matters. In the next section several examples are presented which I believe indicate how some of the technological art of rhelogy has been tranformed through applications of science thereby increasing our capability for predicting a priori how a given material will respond in its flow behavior to certain imposed boundary conditions. Following that I shall attempt to show, again by example, some of the challenges currently offered to the science of rheology by the art. Finally, I close with a few words about the interdisciplinary nature of the subject and a renewed plea for recognition of the interdependence of the fundamental and applied sectors of technology.

II. CONTRIBUTIONS OF THE SCIENCE TO THE ART

For illustrative purposes I wish to describe how applications of basic science have had an enormous impact upon some extremely important engineering problems dealing with flow of non-Newtonian materials, such as polymer melts or solutions.

A. Simple Viscometry

We consider here the matter of interpretation of shear stress—shear rate measurements in a simple viscometer. The resulting rheograms, the term often given to a graphical display of shear stress — shear rate data, constitute the core of information required for any pipeline design of non-Newtonian flow systems. These rheological data are fundamental to decision on pump sizing, viscous heating, and many other important engineering problems.

Suppose that we are interested in ascertaining the ratio (stress/strain rate) for a material for

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which we know there is some unique relationship between τ and strain rate $\frac{1}{\gamma}$

$$\mu_{\text{eff}} = \tau/\dot{\gamma} \tag{1}$$

The classic texebook illustration of this experiment is of course an apparatus in which plane Couette flow is achieved. From the speed of one plate with respect to the other one determines the shear rate. The shearing force per unit area exerted by the plate on the fluid is τ , and μ_{eff} is readily determined as a function of shear rate.

As so often happens, the ideal experiment cannot be the real experiment, and one ends up with a different configuration than that described above. The fluid may be sheared between parallel disks, coaxial cylinders, cone and plate, or by being forced through a slit or circular tube. Two questions arise:

(1) Is the value of μ_{eff} , determined in, say, a plane Couette flow experiment equivalent to the ratio of stress to strain rate in these other flows, several of which possess shear fields which vary spatially over the flow?

(2) If there is an equivalence of $\mu_{eff}(\dot{\gamma})$ in these flow fields, how does one determine $\mu_{eff}(\dot{\gamma})$ in flow fields for which $\dot{\gamma} = \dot{\gamma}(r)$?

Consider tube flow as an example. Pressure change $\triangle P = P_1 - P_2$ is measured for steady laminar flow over length L. From a sample force balance one finds

$$\tau = \frac{\Delta P}{L} \frac{r}{2}$$
(2)

Also, since we can measure volumetric flow rate

$$Q = \int_0^a 2\pi r v dr = - \int_0^a \pi r^2 \frac{dv}{dr} dr$$
(3)

where we have integrated by parts.

Since

$$\dot{\gamma} = \frac{dv}{dr} = \tau/\mu_{eff}$$
 (4)

$$Q = -\int_{0}^{a} \pi \frac{r^{2}\tau}{\mu_{eff}} dr = -\int_{0}^{\tau} \frac{1}{\mu_{eff}} \pi \left(\frac{2L}{\Delta P}\right)^{3} \tau^{3} d\tau$$
(5)

The measured variables are Q and $\triangle P$ for a given fluid in a tube with radius *a*. At this point early workers in rheology were confronted with a dilemma. The desired quantity is $\mu_{eff}(\dot{y})$, but since it varies with position there is no obvious way to obtain it from Eq. (5) unless of course one first specifies some form for μ_{eff} as, for example, the well known power-law relation

$$\mu_{\text{eff}} = K |\dot{\gamma}|^{n-1}$$
(6)

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Many years ago, however, it was found that differentiation of Eq. (5) would achieve the desired result. By using Eq. (2) to change the variable of integration and then applying Leibnitz's rule for differentiation under the integral sign one obtains

$$\frac{d[Q(\Delta P/L)^{3}]}{d(\Delta P/L)} = \frac{\pi a^{4}}{L} \left(\frac{\Delta P}{L}\right)^{3} \frac{1}{(\mu_{eff})_{r=a}}$$
(7)

which is customarily referred to as the Rabinowitsch equation.^{1,*} Although the early rheologists solved this problem by an ingenious *ad hoc* technique, they were really employing a method for solution of integral equations. Recognition of this fact has allowed one to determine more systematically those classes of viscometry problems which are amenable to a simple inversion and those which are not³⁻⁵. Here is an example where knowledge of what might be considered an unduly esoteric subject for an engineer has had an impact upon an important area of applied viscometry, which in turn is fundamental to engineering design of non-Newtonian flow systems.

B. Fluid Characterization

To be sure, the viscometry that we considered above is also fluid characterization. However, I now wish to speak in more general terms about the subject. In dealing with an incompressible Newtonian fluid of known density it is well known that once the viscosity, along with its pressure and temperature dependence, have been determined, the flow behavior of the material is, in principle, ascertainable for any flow geometry; i.e., the coefficients which appear in the Navier-Stokes equation of motion are known. It is also known that the hallmark of a non-Newtonian fluid is the lack of this simple means for characterization. We spoke earlier about necessity of rheograms for engineering design. However it is now known that other information, such as normal stress data, can be informative in providing measures of fluid properties which affect flow behavior. One of the basic questions in rheology is this: What experiments need to be done in the laboratory to characterize the flow behavior of a given material? The answer to this question is strongly dependent upon the com-

*Professor J. L. White once informed me that this procedure was first applied by Herzog and Weissenberg.²

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plexity of the flows to which the fluid is subjected. If we are willing to restrict ourselves to sufficiently uncomplicated flows, then we can say quite a bit about the characterization necessary for very general materials. Probably the best example is the remarkable generality which results through the combination of simple fluids in viscometric flows.^{6,7} In this paper we merely state some of the results which have been obtained from formal application of principles of continuum mechanics.

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We begin by defining a simple fluid as an incompressible material which possesses no inherent anisotropy and for which the stress is determined by the history, up to and including the present, of the deformation gradient. The deformation gradient is a tensor quantity which is a measure of the change in relative position of two neighboring points in a material as it undergoes a motion.⁶ Some reflection will show that the definition of a simple fluid is very general, and, is limited chiefly by the assumption of isotropy and validity of a principle of local action. In essence local action means that only the deformation gradient at a material point of interest is relevant, and one need not consider the effect of the deformation gradient at neighboring points on the point in question. Also needed is a statement to the effect that the response of a body is not affected by rigidbody translations or rotations or, put alternatively, that the response of the body is not affected by motion of the observer. This is sometimes called the principle of material objectivity.^{8,9} Now these elements of themselves do not permit one to say much in a predictive way about fluid behavior. What is required is a linking of this general constitutive behavior with a special class of flows which we shall call "viscometric flows". For purposes of this lecture we interpret viscometric flows as those of the usual viscometers. A common feature is that the flow is characterized by three orthogonal coordinates, the axes being aligned so that the fluid velocity is of the form $\mathbf{v} =$ $\{v(x_2), 0, 0\}$. In fact this is an overly restrictive definition of a viscometric flow,⁶ but it will serve our immediate needs. With these ingredients one can show how three material functions, measured in any one viscometric flow, characterize the flow behavior of a simple fluid in any viscometric flow. The three material functions are

$$\tau(\dot{\gamma}), N_{1}(\dot{\gamma}), N_{2}(\dot{\gamma}) \text{ where} \tau_{12} = \tau(\dot{\gamma}) \tau_{11} - \tau_{22} = N_{1}(\dot{\gamma}) \tau_{22} - \tau_{33} = N_{2}(\dot{\gamma})$$
(8)

and the τ_{ij} are components of the extra stress tensor with respect to the coordinate system used above to define components of v. Definition of the stress in an incompressible fluid always causes some difficulty since the stress is only deter-

minable to within an arbitrary isotropic part. This fact permits one to define τ so that

$$\operatorname{tr} \tau = \tau_{11} + \tau_{22} + \tau_{33} = 0 \tag{9}$$

Then, since the stress is symmetric, the state of stress is completely determined by τ , N₁, and N₂. Furthermore, one can show formally, and in accord with physical expectations, that $\tau(\gamma)$ is an odd function of its argument while N₁ and N₂ are even functions.

Full appreciation of this result requires some knowledge of the vast variety of measurements and constitutive laws with which the literature of rheology abounds. Equations (8) provide a simple means for determining which experiments are equivalent and which are not. Hence one can ascertain the degree to which τ , N₁, and N₂ are rigorously transportable from one flow to another.

The results of Coleman and Noll can be obtained in more than one way. Several years before their publications, Oldroyd¹⁰ used a different argument which led to the same result.¹¹ However, in that paper Oldroyd did not develop the meaning and utility of the three material functions to the extent achieved by Coleman and Noll. It has also been shown that many of the resuls of Coleman and Noll are implicit in some simple symmetry conditions.¹²

In arriving at the results for simple fluids in viscometric flows one again sees how an abstract structure for fluid behavior has given rise to consequences which are of great importance to the practical rheologist and hence to the engineer. The results are significant in both a positive and negative sense. They unify a number of viscometric tests, but also they illustrate that, without further assumptions, little can be said of the generalization to nonviscometric flows.

It is natural to ask whether one can expect

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simple fluids to exist in reality as well as by postulate. Because of the difficulty associated with normal stress measurements this is a difficult question to answer unequivocally. However, limited information about the behavior of polyisobutylene solutions has been consistent with simple fluid predictions.^{13,14}

In connection with measurement of normal stresses it is appropriate to touch briefly upon a remarkable association between normal stress measurement and hydrodynamic stability of rheologically complex fluids. Persons knowledgeable in fluid mechanics are aware of the substantial difficulties that have attended our attempts to understand stability phenomena with Newtonian fluids. This alone might be enough to dissuade one from adding the further complication of nontrivial rheological behavior. However, the return may be well worth the investment. Not only does one find whole new classes of stability phenomena emerging from the presence of elastic response in the fluid, but it seems reasonable to believe that, because of the sensitivity of the stability behavior to normal stresses, stability experiments can be used to measure normal stress differences.^{15,16} This is especially true for N₂ which has proved particularly difficult to measure by more conventional means.

As a brief example of this sensitivity I cite an analysis which has recently been published.¹⁷ We have considered combined momentum and heat transfer characteristics of the flow shown in Figure 1, where a buoyancy force has been superposed upon plane Couette flow by maintaining the lower plate at a higher constant temperature T_1 than the upper plate, which is maintained at T_2 . It is well known from theory and experiment with Newtonian fluids that heat transfer from the lower to the upper plate will occur by conduction until, at some critical condition, the buoyancy force tending to cause con-



Fig. 1.—Boundary conditions for plane Couette flow with superposed temperature gradient.

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vective motion overcomes the counteracting effects of viscosity and thermal diffusivity. This balance is reflected in a critical value of the Rayleigh number, $Ra_c = 1708$, where

Ra = $g\alpha\beta d^4/(\kappa v)$ and g = acceleration due to gravity, α = volume coefficient of expansion, β = temperature gradient, d = distance between plates, κ = thermal diffusivity, and v = kinematic viscosity.

For certain viscoelastic fluids one finds that the critical Rayleigh number is very sensitive to the second normal stress difference N_2 (see Figure 2). This interaction between normal stress behavior and heat transfer characteristics has important engineering implications. Additionally, another indirect method appears for measurement of N_2 .



Fig. 2.—Effect of second normal stress difference (N₂) on critical Rayleigh number (Ra_c). S_{xy} is the shear stress. Reynolds number (Re) and Rayleigh number are suitably defined for the non-Newtonian fluids being considered.¹⁷

Thus, we see again how practical results can be obtained from complicated and seemingly esoteric analysis *if* the persons who are knowledgeable about the basic engineering science also have in mind the needs of technology.

C. Extrusion

As a final example of relevance of advanced engineering science to practical questions, I have chosen a subject that is still far from fully developed. The industrially important operation of extrusion is one of the most complicated of all problems in transport. A polymer simultaneously undergoes phase change, temperature change, and is subjected to wide variations in stress. Even if the constitutive behavior of the material were completely known, the process is so complex that exact solution of the boundaryvalue problem would probably not be possible. However, availability of large computers has made it possible to study the importance of large numbers of variables on the extrusion process, and operating regions have been found over which the effects of different dimensionless groups predominate.



It is convenient to restrict ourselves to operation of a single-screw extruder. Solid material enters one end of the screw apparatus as shown in cross section in Figure 3. In the transition zone the polymer is melted, with accompanying heat effects, and molten highly viscous polymer is transported through the metering section during which there may be strong coupling between momentum — and energy-transfer aspects of the problem because of viscous heat generation. Our discussion will be limited to the metering zone, and we shall see that here alone there is more than ample complexity to challenge both the fundamental research engineer and the persons responsible for choosing an extruder for a particular processing operation.

My remarks are based primarily upon the work done by Pearson and coworkers.¹⁸⁻²⁰ Flow in an extruder is, first of all, certainly nonviscometric. Fortunately, however, it seems that some progress can be made by treating the fluid as a purely viscous *inelastic* material. That is to say, we entirely neglect any effects of fluid memory. This is of course not correct for the typical highly elastic polymer melt subjected to extrusion. Yet, to the order of approximation currently appropriate, the assumption does seem to be useful. The power-law model, which dismisses any effects of memory, has been used.

The first step is to write the relevant differential equations, cast them in dimensionless form, and examine the various dimensionless groups which appear in the governing equations. The number of dimensionless groups, even with the drastic simplification already made to the rheology, is too large to be manageable. However, it is useful to consider the physical significance of various groups and the simplifications which obtain when some of them are restricted in their range. Pearson²⁰ has singled out five dimensionless groups for consideration. These are:

Griffith number =
$$G = b\mu_{eff} v^2/k$$

Brinkman number = $Br = \mu_{eff} v^2/(k\Delta\theta)$
Peclet number = $Pe = Vh/\kappa$
Graetz number = $Gz = Vh^2/(\kappa L)$
Aspect ratio = $A = w/h$

Where b = temperature coefficient of μ_{eff} , V = characteristic circumferential velocity based, for example, on rotational speed of screw, $\mathbf{k} =$ thermal conductivity, $\Delta \theta =$ characteristic temperature difference between walls and melt, and $\mathbf{L} =$ characteristic length scale along fluid streamlines. Geometric factors are shown in Figure 4a.



Fig. 4.—Screw extruder. (a) Geometric factors. (b) Secondary flow.

The values of these dimensionless groups are crucial factors which control the validity of various simplifying assumptions. For example, Pearson notes that the Graetz number, a measure of the relative importance of heat conduction across streamlines to heat convection along them, can vary between 10^{-1} and 10^4 . It is also of interest to point out that in these highly viscous flows the Reynolds number does not appear as a separate parameter.

Though useful in the form given above it appears that, inevitably, a more complicated and thorough analysis should be done. We cite two additional effects on which some work has already been reported.

(1) It now seems quite probable that secondary flow may have a substantial effect upon transport. Consequently, even with such a simple rheological description as the power law, one must recognize that the effective viscosity should be expressed in terms of a combination of the nonzero components of the rate of deformation, namely, the second invariant of the rate of strain tensor.

$$I_{2} = \sum_{i,j} \frac{1}{4} \left(\frac{\partial u_{i}}{\partial x_{j}} + \frac{\partial u_{j}}{\partial x_{i}} \right)^{2}$$

Martin¹⁹ has conducted a numerical analysis using the second invariant. The result is a complicated secondary flow pattern in which there is overturning within a given channel as well as leakage through the flight clearance separating channels (see Figure 4b). Though this secondary

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flow may be a small portion of the total momentum transport it clearly has a major effect on the heat transfer portion of the problem.

(2) Our knowledge of heat transfer in polymer melts under shear is still primitive. To date it is customary to use Fourier's law of heat conduction and a scalar value for the thermal conductivity. However, it is entirely reasonable to expect k to exhibit directional behavior as the polymer is aligned during the shearing process. Indeed, measurements of this effect have recently been reported.²¹

III. CHALLENGES OF THE ART TO THE SCIENCE

In the previous section the tone has been optimistic. I have noted a few cases where application of highly sophisticated research tools, analytical or numerical, has led to a deeper understanding of flow of rheologically complex fluids, and this increase in understanding has had important practical consequences. Now I wish to be more pessimistic in outlook, and to consider some results which show how much remains to be understood about rheology. To do this one need only consider the subject of *non*viscometric flows. We spoke earlier about the great generality in our understanding of the measurements necessary to characterize fluid behavior in viscometric flows. However, the engineer faced with processing problems can argue that flows likely to be of engineering interest will be nonviscometric. Furthermore, suppose that after all of the effort devoted by many people to measurement of τ , N_1 and N_2 , we finally had unequivocal means for making such measurements. What does that tell one about nonviscometric flows? According to general continuum mechanics it tells one very little. However, by postulating certain specific kinds of constitutive equations for a material or by assuming the material to behave according to a particular fluid model — one can learn quite a bit from τ , N₁ and N₂. Here is where the art of rheology must be recognized. It is important to learn which kinds of constitutive simplifications are appropriate for which fluids in which flows. The only way that this can be done is through collection of meaningful data in a variety of nonviscometric flows with a variety of fluids. Bird and coworkers^{22,23} have expended much effort in this direction. From a somewhat different point of view Metzner and his students²⁴ have done experiments with a similar goal in mind. The Delaware workers have done much to show how a simple constitutive equation, the

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convected Maxwell model

$$\underset{\sim}{\tau} + \theta_{f\ell} \frac{\delta}{\delta t} \underset{\sim}{\tau} = 2\mu \overset{\bullet}{\chi}$$
 (10)

gives rise to an ordering parameter which is a useful guage of fluid response. Here θ_{f1} is a characteristic relaxation time for the fluid and $\delta/\delta t$ represents a convected time derivative taken with respect to a coordinate system that is translating and rotating with the fluid. If one uses a time scale θ_p as the time over which an element in the flow process undergoes an appreciable change in, for example, strain rate, then the ratio θ_{f1}/θ_p is an important parameter. The magnitude of this ratio, often called the Deborah number, indicates whether the material will exhibit primarily fluid — or solid-like behavior.

I wish to return briefly to hydrodynamic stability. In the previous section the subject was used as a source for examples of "contributions of the science to the art". However, stability of non-Newtonian fluids is still in an early stage of development, and all of the theoretical work cited earlier rests upon some important simplifications. We are still groping for a clear statement of the constitutive properties which govern stability behavior. Once a laminar shearing flow has become unstable it is of course no longer viscometric. In fact since stability analysis is performed by computing the behavior in time of disturbances superposed upon the basic viscometric flow, one can argue that the disturbed flow is no longer a viscometric flow. Goddard and Miller²⁵ and Pipkin and Owen²⁶ have produced analysis in which they show with some rigor how one proceeds to treat small departures from viscometric flow. They show that in general one needs several new material functions to describe small departures from viscometric flows. Furthermore, it is not at all clear how one would proceed to measure these functions. There are two ways to avoid this problem. One is to choose a specific constitutive model which is also simple enough to permit linear or nonlinear stability analysis to be performed.²⁷⁻³⁰ The other is to neglect what one hopes are terms of second-order smallness and to proceed with a linearized stability treatment.^{15,17} Although further work is needed, there do seem to be experimental indications that the latter treatment has some validity.¹⁶

Before leaving the subject, I wish to note two examples of current interest which demonstrate our lack of understanding of hydrodynamic stability of viscoelastic fluids:

(1) In the flow between coaxial cylinders there are indications that instabilities can exist at **very** low rotational speeds. These have been postulated to be transient effects,³¹ but that explanation is not yet a certainty.

(2) Secondary flow phenomena associated with free surfaces appear to be far more complicated than originally supposed. In particular, Saville and Thompson have found secondary flow cells associated with the Weissenberg climbing effect.³² Work is underway at Princeton which is aimed at a more detailed study of these and other secondary flows.

A talk dealing with current topics in rheology would not be complete without mention of the popular subject of drag reduction. It seems clear that the action of drag reducing agents is due to an interaction between the turbulent eddy structure and the macromolecules added to the system. However, the precise nature of this interaction is still unclear. As the review by Lumley³³ emphasizes, the proper roles of length and of time scales are not yet understood. This fact notwithstanding, an impressive correlation based primarily upon length scales has recently been proposed by Virk.³⁴

IV. RHEOLOGY AS AN EXAMPLE OF AN INTERDISCIPLINARY SUBJECT

From the examples already given it is apparent that the subject of rheology is a meeting ground for applied mathematician, physicist, chemist and engineer. Consequently one can easily observe both the powers and the pitfalls of interdisciplinary research. This is especially true with the subject of suspension rheology, a final example on which I wish to comment.

Almost all treatments of suspension rheology begin with proper hereditary acknowledgment to Einstein,³⁵ who developed the famous expression for the viscosity of a dilute suspension of rigid spheres in a Newtonian medium. Physical chemists have shown a continuing interest in the subject because of the utility of a suspension model as a means for understanding flow behavior of macromolecular systems.³⁶⁻³⁸ The subject also has appeal to those interested in flow of fluids past rigid and deformable bodies, since most developments in the rheology of suspensions begin with a solution for the velocity past a body placed in a shear field.^{39.41} Recently there has been an interest in the behavior of DNA and other biopolymers in shear fields.⁴² It appears that through such studies we can gain useful information about the conformation and flexibility of such molecules.

The advantages of bringing a multiplicity of backgrounds to bear on a problem are obvious. However, a price has to be paid. First of all the task of maintaining an awareness of current research is greatly complicated. When it is equally likely that a given problem may be discussed in *Biopolymers* or the *Journal of Fluid Mechanics*, not to mention about twenty other journals bounded by those extremes, the individual researcher is faced with a difficult task of information collection. This is compounded by the fact that it is even more important to be aware of *current* work in various parts of the world, as opposed to the completed projects which are reported in journals.

A second difficulty is the gap in vocabulary and approach that exists, particularly between those trained in biological or polymer science and those trained in applied mathematics. Useful interchange requires a substantial degree of effort and patience from all concerned.

In spite of these difficulties, which are certainly to be expected, the possible benefits from such a broad variety of backgrounds are substantial and have, I think, been amply demonstrated.

V. CONCLUSION

From this tour through some of the knowns and unknowns of rheology I hope that a case has been made for the practical utility of fundamental engineering research, provided that those who do it are aware of the needs of the applier. That applier may, in the present instance, be a polymer processing company, a government laboratory, an equipment manufacturer, or even a hospital or a medical research team. One of the claims implicit in fundamental research (if it is engineering research) is that it will have application to a *number* of specific needs. This fact is worth remembering, particularly in universities as the pressure builds for mission-oriented efforts by teams of faculty and students. Good fundamental engineering research need not, and perhaps should not, be done in the absence of any specific need. However the result, if it is of high quality, will transcend the particular need for which the work was begun.

Though the activity of an industrial research engineer is expected to be closer to specific application than that of his academic counterpart, one should not forget that an important function is to screen the work of others for possible utility. To do this the industrial researcher needs the advanced training in engineering and science that will enable him to understand the contributions of others and to recognize those which can be made useful.

All of these pleas have been made before, but they need to be made again. In the current economic climate both the researcher and the applier need all the help that they can get. I suggest that they renew their efforts to help each other. \Box

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