

## EPILOGUE TO THE NATO ASI ON FUNDAMENTALS OF FRICTION

### 1. Introduction

The NATO ASI at Braunlage was a fruitful exploration of the current state of knowledge and new approaches to friction processes (and other tribology-related matters). In addition to formal presentations, most of which are included in this book, the ASI devoted much of the last two days to discussing what areas of friction are well-understood and what issues need further clarification. The epilogue, divided into 5 parts, is a summary of those discussions as well as more recent inputs. The last day of the meeting was devoted to questions, answers and commentaries on "Future Issues in Friction at the Microscopic and Macroscopic Level." One of the commentaries, by David Tabor, nicely captures the atmosphere of the meeting and provides a clear and concise summary of many of the issues addressed. A transcribed version of his comments is given in part 2, below.

In preparation for the "Future Issues" session, the penultimate day was devoted to topical discussions. Eleven groups were organized, and each was asked to identify:

- What concepts of friction have been made clear on the macroscopic and microscopic level?
- What is missing (concepts, theoretical or experimental approaches)?
- What topics were not treated (fully) that need to be dealt with in the future?

Groups were kept smaller than fifteen, and participants were asked to wander from group to group and contribute to topics of interest. At the end of the session, the participants reconvened to hear 5-minute summaries (later transcribed and printed) from each group leader. The summaries and transcribed comments from the "Future Issues" session were distilled by the editors into a list of **ISSUES and RECOMMENDATIONS**. These lists (**printed in bold letters**), along with editorial comments and selected commentaries (*in italics*), are given in part 3. Two of the topics that generated much discussion, "Energy dissipation by friction processes" and "New ways of probing friction processes," are given extended attention.

Finally, everyone loved the molecular dynamic simulation videos of Uzi Landman, Jim Belak and Judith Harrison and the BBC science program with Thomas Mathia on "tribology of skiing." While there is no substitute for a good movie, a good diagram is sometimes second best. Lots of creative effort was spent generating diagrams depicting "Hierarchy of Friction Models" that span the microscopic to macroscopic. Three of these are presented in part 4. We end the epilogue in part 5 with the final comments of a lubrication engineer, Duncan Dowson, who wonders out loud how this knowledge of "microscopic friction processes" might influence engineers.

### 2. Commentary by David Tabor.

*"This Conference I think has been for me the most exciting and worthwhile Conference on Friction that I have ever attended. I have been greatly stimulated, and I think all of us have been, by the lectures, by the attention of the attendees, by the discussions, by the posters, by the participation of the audience as well as the lecturers and by the flexibility in approach shown by all our specialist participants. I would like to thank the organisers and the organising committee on my behalf and on behalf of all of you for arranging such a successful event.*

*The continuum approach. In attempting to assess major conclusions it seems to me that there is a consensus that continuum mechanics, contact mechanics and fluid mechanics will continue to supply the practicing engineer with the tools for designing and producing effective and viable machinery. But he or she will need to know more about the properties of materials*

From: *Fundamentals of Friction: Macroscopic and Microscopic Processes*, eds. I.L. Singer and H.M. Pollock, Kluwer, Dordrecht, 1992, pp.569-588.

and the properties of fluids, and this remains an ongoing problem for the tribologist. From the practical point of view bulk properties will be sufficient but when, for example, the EHL film reaches thicknesses of the order of a few hundred angstroms (as indicated by Duncan Dowson) we may have to consider in greater detail the molecular structure of the liquid. I shall refer to this later.

Surface topography. In this continuum approach to friction and lubrication we still have to struggle with the problem of surface topography. Probably the design engineer will continue to use the elegant profilometry techniques described twenty five years ago by Greenwood and Williamson - in spite of what Jim Greenwood tells us about some fundamental unresolved conceptual difficulties. I feel in the discussion of this issue like the happily married couple who after 25 years of married felicity suddenly learn that their marriage certificate is faulty and may be legally invalid. Nothing in their relationship is changed but they are not quite sure where they are. My impression is that engineering tribologists are well and truly wedded to the established ideas of surface profiles, distribution of asperity heights, radii of curvature of asperity tips and sampling distances. For the foreseeable future the old relationship will remain as a fundamental constituent of tribological design. It is up to those who have discovered a flaw in the marriage certificate to rectify the situation. I am sure we all wish Jim Greenwood and his colleagues every success in this serious task of preventing a divorce.

Energy dissipation. When we talk about friction we nearly always think of it as a force. It is very rare that workers talk of energy dissipation and when they do they refer to it in terms of bulk properties e.g. plastic flow, viscoelastic losses, viscous flow. And I think it is at this point that I notice a change in attitude at least amongst those who are interested in the mechanisms of friction. We are beginning to recognise that maybe we should be thinking of energy dissipation in terms of atomistic processes. For example the talk I gave here is not a lecture I took out of my file of lectures: it was one I had never delivered before because I had not been thinking specifically about energy dissipation, except in terms of bulk properties. It was stimulated by a questionnaire sent out by the Conference organisers. My approach seems very simple and obvious. I started with the friction of metals where it is known that plastic deformation usually occurs at the regions of real contact. We know that plastic deformation involves unstable displacements by unit atomic spacings along the slip plane, the atoms vibrating as they jump from one position to another. I came to see that all frictional processes - or most of them - involve a distortion of the atomic arrangement, an instability, a flicking back to a new position of equilibrium while the distorted material vibrates and the vibrations ultimately degrade into heat.

Of course with metals we may still think in terms of plastic deformation and simply assert that there is no problem here since it obviously involves the expenditure of energy. Similarly with viscoelastic solids we may explain energy loss in terms of springs and dashpots. But polymers are not made of springs and dashpots. They are made of molecules which in themselves have intrinsic elastic properties of rate-dependent modulus and they are impeded in their movement by molecular interactions or entanglements which involve viscous losses.

Surface films. You can see from what I have just said that I have been thinking of friction in terms of bulk properties interpreted in molecular terms. I have not said a word about surface films which in the real world play such a significant role in the frictional process. Sometimes these films are applied directly to the surface or are incorporated into the surface during manufacture. But in many cases they are formed by chemical reaction between the surface and the environment. For this reason I welcome, as one of the important aspects of our lectures and discussions, our exposure to the chemistry of film formation. Of course with engineering systems

many of these surface reactions are facilitated by the temperatures developed by frictional heating and I hope that at some stage we shall have a more detailed understanding of the temperatures developed between sliding surfaces and their role in chemical reactivity. A day or two ago we heard that, according to molecular simulation calculations, surface temperatures of three or four thousand degrees can be achieved and we need to look more deeply at this (see later).

Apart from the way surface films are formed we need to know how well they are attached to the substrate, whether they are ductile or brittle and how we can quantify these properties. So far we can only refer to them in descriptive terms.

Surface proximity devices. In referring to surfaces and surface films this brings me naturally to that part of the Conference which has dealt with the surface-force microscope (SFM) and I am glad that some effort is now being made (as we heard from previous speakers) to enlarge the area of surface that can be usefully examined by a modification of the SFM technique. This may bridge the gap between atom-by-atom studies and the somewhat more macroscopic aspects of friction which have been studied so elegantly by Jacob Israelachvili and Steve Granick.

Molecular dynamics: atomic simulations. Here we come I think to what I regard as one of the fundamental problems involved in our understanding of friction: that is we do not have a way of seeing what is actually taking place at the interface while sliding is taking place. We can look at the surface before and after sliding has occurred. But what took place at the interface is inaccessible to direct examination. There are of course some exceptions. If one surface is transparent we can study the hot spot temperatures from the radiation emitted but the details of the contact regions remain unknown. And that is why the beautiful atomic simulations by Uzi Landman and Judith Harrison present such an exciting prospect. I was in fact familiar with some of the earlier work in molecular dynamics especially the classical pioneering studies of Alder but at that time it did not seem to me that this could offer a useful approach to friction simply because so few atoms (or molecules) were involved whereas the individual junctions in friction experiments, even at loads of micronewtons, contain thousands or millions of atoms. Indeed it has always been my view, in the past, that atomic models of friction are "unrealistic" and in some ways I am surprised at my own lecture with which I opened this Conference. But we now hear that modern computations make it possible to study the behaviour of 10,000 atoms, say a cube of 20 x 20 x 20 atoms and that these simulate, to a remarkable degree, the behaviour of bulk matter.

Seeing that modern molecular simulation has been so successful I would like to hope that further work along the lines of Evans in Australia will be carried out by tribologists on the viscous properties of liquids, in particular on the effect of temperature, pressure and shear rate. Perhaps too such studies will clarify the boundary conditions which determine whether there is any slip between the liquid and a solid surface.

Surface temperatures and surface films. In connection with molecular simulation I would like to repeat two items that I mentioned earlier in relation to the frictional process. We can calculate surface temperatures in friction from a knowledge of the frictional work, the speed, the thermal conductivity of the solids and from a reasonable estimate of the asperity size. But Uzi Landman has suggested that very much higher temperatures (not yet thermodynamically equilibrated) may be generated in the brief instant that transpires as atoms slide over one another. It is important for us to know if these are real or significant in affecting surface properties and surface reactivity. This certainly merits further study. The second concerns the

*behaviour of surface films during sliding whether they are organic lubricants or solid films. Can molecular simulation help us to understand their mechanical properties and how they break down? In practical affairs these films are of great importance.*

*A new springtime? This is the first time I have been exposed to a detailed account of the power of molecular (or atomic) simulation in the study of surface interactions. I do not know if the details depend crucially on the assumed interatomic potentials but I think this technique offers great promise for further work in this field. When as a physicist I began my research on friction over 50 years ago our surface techniques were limited to the optical microscope and a little later to electron microscopy and diffraction. At that time we could at best interpret our results in terms of bulk properties. Almost every experiment that we did was being done for the first time and I suppose I could say that the first 15 or 20 years of this work constituted the spring time of our work in terms of our macroscopic approach. I think we are now approaching another spring time (as Ernest Rabinowicz indicated in his opening talk) and that this will involve a critical and constructive application of molecular simulation to the frictional process. It is not possible to tell if such an approach will lead to better tribological practice but I am sure it will shed new light on the mechanisms involved.*

*There is of course a great danger. I can imagine that if in three or four years' time we have another Conference on Friction we shall have nothing but papers on the atomic force microscope and molecular simulation. Meanwhile in the background the practical people will be studying the wear properties of surface films (especially plasma deposited diamond), the degradation of organic lubricants and other mundane matters - while our engineering tribologists, like Duncan Dowson and Ken Johnson will be busily engaged designing more efficient and more viable machinery on the basis of continuum properties. And Ernest Rabinowicz will be measuring the slipperiness of floors and earning the just rewards that flow from the American obsession with litigation.*

*This has been a most exciting Conference. I hope that you will not feel more tired than I am and that you will return to your laboratories stimulated by the papers, the discussions and the camaraderie of the last ten days.*

### **3. Issues and Recommendations.**

#### **3.1 ENGINEERING MACHINE DESIGN**

- 1. How can engineers make quantitative predictions of machine behavior from present-day models of friction behavior (continuum mechanics, lubrication, contact vibrations)?**
- 2. Can we characterize the boundary/interface conditions of machines well enough to apply friction models?**
- 3. What friction-related parameters can be used to monitor vibration and vibration instabilities?**
- 4. How can engineers distinguish and cure or control the different forms of stick-slips?**
- 5. Can machine motion be tailored to optimize transfer films for low wear and stable friction?**
- 6. Microscopic engineering: How might fine-tipped proximal probes influence the**

design of future engineering devices, from the submicrometer to sub millimeter scale? They can be used to manipulate materials, cut patterns, punch holes, polish asperities,...

7. How will engineers cope with the intrinsic "high friction" of micromachines, given that they will likely be fabricated from "clean" materials (e.g. Si) and will likely be operated in "clean" environments and at relatively low loads?
8. Can engineers take advantage of the "new found" properties of confined fluids (<10 nm thick) to control friction?
9. How can properties of ultra-thin lubricant films be incorporated into lubrication models to predict behavior of ultra-smooth engineering surfaces?

### 3.2 DEFICIENCIES IN PRESENT-DAY MACROSCOPIC MODELS OF FRICTION.

1. Models are relatively well developed for idealized materials subjected to elastic, visco-elastic or simple two-dimensional plastic deformation.
2. Models are needed to account for: strongly layered thin films, work hardening, and brittle crack growth under compression in idealized materials and in real materials (crystal structure, grain size, dislocation behavior,...).
3. How does friction evolve with sliding? Can "dynamic friction maps" be constructed analogous to "static friction maps?" [see chapter by Childs and extended discussion by Johnson.]
4. At what separation do continuum models (e.g. elastic/plastic deformation, roughness,...) breakdown?
5. What are the mechanics of interfacial wave propagation?
6. How does friction enhance the stress intensity field in mode II fracture?
7. What role does fracture mechanics play in sliding motion and attendant frictional resistance? Is there mixed-mode failure? This question follows from the presentation of Johnson (p. 227) in which he showed how certain materials fit onto a friction "map" whose different regions represent steady sliding, brittle stick-slip and ductile plastic behavior. During the discussion that followed, Arvin Savkoor made the point that the possibility of mixed-mode failure is still an open question. The relevant issues here are:
  - a. what are the alternative "rival" processes (e.g. mode I-type Schallamach waves for elastomers, constrained plastic failure for metals,...);
  - b. given that the normal and transverse stress distributions determine  $K_I$  and  $K_{II}$  respectively, how do these combine to give the total work of adhesion? Once this is known, it should be possible to predict the condition for instability (slip);
  - c. if mode II fracture is operating, do we assume that fracture energy is *not* recovered through healing of the crack? [This question is quite separate from the question of energy loss mechanisms!];
  - d. is it necessary to postulate an intrinsic shear stress of the interface,  $\tau_0$ ? Can this be related to  $K_{IIc}$ ? If not, must we always postulate the existence of a surface film whenever we need  $\tau_0$ ? [see chapters by Briscoe and Savkoor].
8. How can the models be modified to account for the "intermediate scale

**discreteness" of real materials including dislocation behavior, crystal structure, grain size,..?**

- 9. Can material-dependent models be formulated that account for "intermediate scale discreteness?"** Clearly, in some cases we can bridge this very large gap between the continuum models used by engineers and the models involving discrete atoms. But, as pointed out by many speakers, microstructure gives rise to another type of "discreteness."

In this connection, Johnson asked how we can integrate the activities of people who work on the atomic scale with those on the engineering scale:

*"These remarks have not been carefully thought out and should be regarded as a spontaneous reaction to the meeting which is coming to an end. They relate to length scales and the relation between those who work on the small scale and those on the engineering scale. First of all it has been a splendid occasion for people who work in these two areas to talk to each other and to discover what we are each trying to do: in many cases in different departments.*

*I am an engineer: probably most of the people in the room are physicists and we do not meet as often as we should. So that has made it a splendid meeting. On the other hand, although we now have this feeling of esprit de corps, I think we somehow kid ourselves if we think that we can start with forces between atoms and finish by calculating answers to problems such as shoes gripping floors that Ernie Rabinowicz was talking about. I don't think that one does that sort of thing.*

*One of the things that has struck me is that there is an intermediate length scale which we have not heard much about at this meeting. It lies between the atomic/molecular behavior that we have heard a lot about and the continuum approach. In metals, for example, it is the scale of dislocations and other defects in crystals. It is the scale where crystal structure matters.*

*I think back to when I was a beginner in the tribology game, just about the time that dislocations were 'invented' or at least the time when they were beginning to seriously affect material scientists' thinking. I was terribly worried whether I would have to learn all this very complicated dislocation mechanics in order to perform calculations of the plastic deformation of surfaces. But that isn't the way it is: one doesn't solve plastic deformation problems of an engineering sort by starting off with forces between dislocations.*

*Thanks to this meeting, we engineers now have a whole new area of activity to get used to, one which will surely be important and feed into tribology. But we kid ourselves if we think we can start off with forces between atoms and solve engineering problems on a continuous line carried out by a single person."*

This lead on to the question of asperities:

### **3.3 ROLE OF ASPERITIES IN SLIDING CONTACT.**

- 1. Have friction models properly accounted for material properties at the asperity scale?** In addition to the general points made above, it was pointed out that a serious attempt to correlate observations of frictional behaviour with grain boundary behaviour could be worthwhile. It appears that on the whole, single-asperity fibre/fibre sliding experiments are consistent with macroscopic models -- the gap here is between the asperity scale and the atomic simulations.
- 2. How does the contact in static loading evolve as sliding commences?**

3. **How does the surface roughness evolve during dry sliding contact?**
4. **At what length scale does the flow stress approach the theoretical stress?**
5. **How do fracture and plastic effects compete at the asperity scale?**
6. **Does the wear mode change as the contact size gets smaller?**
7. **Do AFM and fine-point friction studies depict friction at the "asperity" level?** [see chapter by Pollock.] **If so, why are the results inconsistent with results inferred from powder flow friction studies?** [see chapter by Adams.]
  - a. **Perhaps these proximal probe experiments are in the regime that should not be considered the single asperity scale?**
  - b. **What other experiments can be done to understand friction at this scale?**
8. **Does surface energy play an important role in friction processes at the asperity scale?** Brian Briscoe argued that surface energy plays an "intermediate" role - to hold surfaces together long enough to allow strain energy to be transferred to the regions adjacent to the contact. However, the strain energy itself is dissipated into the bulk.

### **3.4 ROLE OF INTERFACIAL SLIDING AND RHEOLOGY.**

1. **Where does sliding occur? Technically, where is the "velocity accommodation locus?"**
  - a. **How does this locus change with separation between the counterfaces?**
  - b. **Where is the microstrain accommodated? At a counterface (by sliding) or within the interfacial material (by shear)?** We must identify the velocity accommodation locus: 1) to advise the microscopic modelers what occurs in practice, and 2) to understand wear, transfer and traction cracking.
2. **What mechanism governs the magnitude of the "velocity accommodation parameter" or "interfacial rheology parameter," commonly called the interfacial shear strength or contamination factor and labeled  $\tau$ ,  $q$ , or  $s$  by various authors?**
  - a. **What is the significance of the velocity accommodation parameter and how is it related to bulk properties?**
  - b. **How do gas-surface reactions alter the velocity accommodation parameter (and wear behavior) of sliding contacts?**
  - c. **How is this parameter related to energy dissipated in solids and fluids?**
3. **Can the magnitude of the velocity accommodation parameter ("shear strength") be predicted from properties of interfacial films or their bulk material counterparts?**
4. **How does the "third body" evolve?**
  - a. **What are the rheological changes and dissipation characteristics?**
  - b. **Are the processes isothermal, because of small volumes involved?**
  - c. **What are the processes (mechanical and chemical) by which frictional energy transforms solid/liquid/gas interactions into surface films and third bodies?**
5. **What is the relationship between friction and the generation of wear debris?**
  - a. **Do frictional stresses control the size, shape, structure or microstructure of wear debris?**
  - b. **If so, what can this analysis tell us about the friction processes that generated**

the particles?

- c. Can this friction-wear synergism be used to control wear?
6. Can wear models be devised that can predict "lubricating" third body products that reduce friction and minimize wear?
7. Are friction processes at the macroscopic and microscopic scales governed by different mechanisms? Interfacial sliding studies in both traditional friction tests and in the SFA show similar friction behavior - either  $F \propto \text{load}$  (Amontons' Law) or  $F \propto \text{area}$ . However, different interpretations of apparently similar behaviors have been given. Amontons' Law is accounted for traditionally, e.g. in the Bowden-Tabor model, by shearing of junctions accompanied by *permanent* plastic deformation. Israelachvili, however, [see p. 371] derives the same behavior using the cobblestone model, in which heat is dissipated with *no permanent* deformation. Moreover, the friction coefficient in the two cases is determined by very different materials properties [see p. 372]. The second behavior ( $F \propto \text{area}$ ) leads to two different explanations of the interfacial shear strength,  $\tau = \tau_0 + \alpha$ . Briscoe [see p. 167] interprets  $\tau$  (generically) as a dissipation parameter and  $\alpha$  as the pressure dependence of the shear strength (of the "third body" or the interfacial itself); Israelachvili [see p. 371] suggests  $\tau_0$  is due to attractive van der Waals forces and the  $\alpha$  term is the "bumpiness" of the contact.

### 3.5 SURFACE FILMS AND ATOMIC LEVEL TRIBOCHEMISTRY.

1. If we want to control friction and we have the ability to precisely modify the chemistry and structure of surfaces, what chemical and physical properties would we give to the first few hundred angstroms of a metal or ceramic?
2. How do surface films accommodate stresses or transmit them to the subsurface (quantitative models)?.
3. Is frictional energy dissipated in surface films? If so, what is the mechanism?
4. How do friction processes activate surface chemistry? Traugott Fischer offered several possible mechanisms:
  - a. By direct mechanical excitation of a chemical reaction, e.g., by straining surface bonds;
  - b. By thermal activation of a local volume of material;
  - c. By removal of reaction products;
  - d. By acceleration of diffusion processes (mass transport);
  - e. By exoemission;
  - f. By triboelectricity.
5. If friction produces the rapid local temperature rises suggested by molecular dynamics simulations, ca. rates of  $10^{10}$  K/sec, how should one describe the chemistry?
  - a. Would it be vastly different than what one would get from a surface held at a constant temperature of that value?
  - b. What kinds of chemistry e.g. the modes of energy dissipation (vibrational, rotational, electronic,...) takes place under these conditions? Perhaps, chemical

dynamic simulations will be necessary to identify the chemistry. This point pertains to issues discussed following the lecture by Fischer, on page 310.

6. **How does crystallinity (lattice imperfections, surface registry, amorphous vs crystalline,...) affect tribochemical reactions?**
7. **How do tribochemical films that form during sliding or rolling contact affect tribological behavior?**
8. **What are the chemical and mechanical contributions to the breakdown of tribochemical films?**
9. **What are the important properties of films, e.g. surface energy, that should be examined experimentally?**

### **3.6 HOW CAN ATOMISTIC MODELING MAKE AN IMPACT ON UNDERSTANDING FRICTION?**

#### **Approaches:**

1. **Identify the microscopic phenomena underlying macroscopic behavior. (topics listed below)**
2. **Describe properties of small material aggregates.**
3. **Assess and evaluate the range (particularly spatial) of validity of continuum descriptions.**
4. **Topics that can be treated include:**
  - a. **Material transfer,**
  - b. **Surface and subsurface diffusion,**
  - c. **Atomistics of plastic response,**
  - d. **Energy dissipation mechanisms such as transient heat generation or excitations (phonons),**
  - e. **Schallamach and Stoneley waves -- generation and propagation,**
  - f. **Phase transformation (amorphization, glassification, solid-liquid, etc.),**
  - g. **Boundary conditions (stick-slip and partial slip),**
  - h. **Non-equilibrium phenomena: Energy pathways and mode coupling,**
  - i. **Mechanical response to transient heat and pressure generation, including shock conditions,**
  - j. **Chemistry under temperature jump (flash) conditions,**
  - k. **Interfacial shear strength, its variation with depth and the tribological consequences,**
  - l. **Tribological consequences of surface modifications (e.g. ion implantation, alloying).**

#### **Problems:**

1. **How critically does the deduced friction process depend on the form of the *assumed* interatomic potential?**
2. **Can algorithms (e.g. hybrid methods) be developed to simulate friction processes at time and length scales longer than can be treated in molecular dynamics calculations alone? e.g. that extend computational simulations from the**

**nm/femtosec scale to the  $\mu\text{m}/\mu\text{sec}$  scale.** It seems that molecular dynamics is unlikely to ever be able to incorporate behavior over all relevant time scales into a single simulation. In a given engineering application, it will always be necessary to couple the molecular dynamics approach with the continuum mechanics approach at the appropriate length scale.

3. **Can atomic models treat particular materials or are they only able to qualitatively distinguish between classes of materials e.g. ionic vs covalent vs metallic?**
4. **In practical machines, the surface films that sustain sliding -- whether organic lubricants, oxides or other solid films -- eventually break down. Can molecular simulations help us to understand the mechanical properties of these films and how the films break down?**

Fischer's group of wandering scholars analyzed how one constructs an atomic theory of sliding friction. Assuming that shear takes place at a solid-solid interface and considering only those mechanisms by which translational movement is transformed into heat, they suggest six building blocks for such a theory:

1. The interatomic force potentials inside the sliding body;
2. The atomic force potential between atoms of the two opposing bodies;
3. The relation between these potentials, to determine whether adiabatic (non-dissipative) sliding is possible or if the interbody bonds 'snap' and cause vibrations of the atoms;
4. The crystal structure of the two solids must be considered, including questions of registry between the two structures;
5. Dynamics (how fast is vibrational energy emitted into body?) and viscoelasticity;
6. Collective effects (sliding by dislocation motion, shear melting, creation of defects).

Finally, Belak's group came up with a strategy for calculating engineering friction coefficients by atomistic methods; their strategy is presented in part 4.2 below.

### **3.7 SQUEEZED LIQUID LAYERS AND BOUNDARY CONDITIONS.**

1. **Can lubricants be tailored to take advantage of the dynamic properties of certain fluids** [see "chemical hysteresis" in chapter by Israelachvili, p. 351, **and extreme non-Newtonian fluid behavior (e.g. enhanced critical shear stress, viscosity, elasticity...)** [ see chapter by Granick, p. 387].
2. **What is the origin of the time-dependent static friction of confined fluids?** Granick, on p. 387, suggests two explanations: growth of the area of contact and slow molecular rearrangements at the boundary.
3. **What atomic scale behavior influences liquid lubrication?**
  - a. **At the boundary, e.g. degree of stick or slip at wall.**
  - b. **In the "squeezed fluid," changes in phase or chemical composition, changes in rheology.**
4. **How do the above changes influence cracking and wear?**

### **3.8 HOW CAN ATOMISTIC MODELING MAKE AN IMPACT ON UNDERSTANDING**

## LUBRICATION?

Computer simulations can be used to study:

1. Effect of molecular structure on phase and rheology,
2. Effect of temperature, pressure and strain rate on phase and rheology,
3. Changes induced by additives or mixtures,
4. Effect of loading rate,
5. Modes of energy dissipation,
6. Non-Newtonian flow,
  - a. Where does shear occur? (at surfaces or in lubricant?)
  - b. How do the molecules move (what is the activation volume)?
  - c. What are the relevant time scales?
  - d. What determines the large shear-rate limit of the shear stress?
7. Artificial quasi-solids as a fast technique for solving continuum equations.

### 3.9 STICK-SLIP PHENOMENA.

1. How should we categorize the many contributions to stick-slip? Should we distinguish engineering phenomena (e.g. thermal softening in brakes, roughness effects on rolling surfaces, incipient seizure on clean surfaces) from friction oscillations or ratcheting at the atomic scale? Is the latter stick-slip?
2. What criteria can be used to distinguish stick-slip from ratcheting?
  - a. length scales? Mark Robbins suggested we call an effect stick-slip if the length of the slip increases as you decrease the spring constant. If you have ratcheting motion, the length of slip is tied to a length scale intrinsic to the underlying substrate. This suggestion could provide a needed criterion for differentiating between true stick-slip and ratcheting over atomic potentials.
  - b. spring stiffness?
    - i. Robbins also indicated that varying the spring stiffness can produce qualitative changes in the relationship between force and velocity.
    - ii. spring stiffness also imposes a fundamental limitation on friction measurements with an AFM: for high sensitivity, the spring must be so weak that it moves almost the characteristic length of the atomic potential variations.
    - iii. an important point is whether or not the  $kT$  energy of the sensing spring used to measure frictional force is as large as the frictional energy that you are trying to detect.

### 3.10 COMMENTARIES ON ENERGY DISSIPATION MODES IN FRICTION PROCESSES.

Tabor's lecture and the discussions that followed (see pp. 3-23) set the scene for subsequent contributions to the understanding of this central topic. On the last day of the meeting, he added the following remarks on the subject of surface energy:

*"Before I attempt to express my main impressions of this Conference I feel I must interpose a few words as a penitent and apologise for the mistake I made at the end of my original*

presentation. On that occasion I presented a model which I thought demonstrated that in pulling two surfaces apart the process was reversible and only the surface energy was involved. I now realise that this is not so and that as the bonds go "pop" across the interface elastic energy is lost in the bodies. I do not know if this is true in peeling. But my original model involving the separation across a plane appears to be wrong.

As a practising scientist I recognise three levels of achievement. The first and best is to get it right and to have the results accepted by others. The second is to get it wrong or to be controversial and to evoke discussion which finally resolves the issue. The third and least satisfactory is to have your work ignored. My involvement in surface energy is in the second category."

#### Other commentaries on energy dissipation:

(John Yates) "It seems to me that the field of friction is just beginning to meld together macroscopic and microscopic pictures. A similar transformation happened at an earlier time in the field of chemical kinetics, where for the previous 75 years, investigators had focused on measuring and explaining rate constants for chemical reactions. Then the dynamicists came in and began to measure the individual trajectories of processes that occurred on the atomic level. The objective of that work **never** was to calculate the overall rate constants from all the microscopic measurements, but instead to do the microscopic physical measurements and the modeling in a way that allows one to see entirely new concepts developing. Such concepts could never come out of studies of rate constants which are simply the global averages of the effects of all of these processes. It's likely, therefore, that the goal of the microscopic approach to friction is to introduce entirely new concepts of friction processes, rather than simply finding a way of calculating friction coefficients or other engineering parameters from more fundamental processes."

(Gary McClelland) "We must develop experimental techniques to learn how energy is dissipated in frictional processes. Following the path taken in chemical physics, instead of simply looking at the rate at which chemical energy is dissipated, we should look at the modes of energy dissipation. In tribology, we should look at the spectrum of energy dissipated in frictional processes. For example, what events generate high frequency phonons and what events generate low frequency sound waves? High frequency phonons, for example, may be generated by short, prompt events. Since these events are very short, you would have to use a spectroscopic technique which captures these rapid events and likely do experiments at low temperature where the lifetimes are much longer."

(John Pethica) "We recognize that friction processes result in heat dissipation. But that doesn't mean that heat was the initial energy loss process. We know, for example, from triboemission studies, that there are high energy processes that occur during rubbing. Therefore, following the chemical kinetics/dynamics physics analogy, perhaps if we sort out what all these spectral responses are initially, we may come to recognize the dominant processes controlling friction and the mechanics of energy dissipation."

(Jacob Israelachvili) "With friction, just as with adhesion, what you always have is a loading followed by an unloading. This may be viewed in terms of an approach then a separation, or an advancing then a receding contact angle. And, regardless of the size of the interaction zone, the front part of a sliding asperity is in the loading (adhering or entangling) state and the back part in the unloading (de-adhering or disentangling) state. The loading and unloading events are, therefore, separated by some characteristic length and time, which clearly

*depends on the sliding velocity.*

*Now let's consider energy dissipation. Dissipation will be governed by two **competing** processes occurring at the front and back of the sliding contact zone. Consider the two extremes of fast and slow sliding velocities. At very high speeds there is no time for entangled or adhesive junctions to form (at the front), while at very low speeds there is plenty of time for separation or disentanglement of junctions to occur at the back. In both cases, there is little departure from equilibrium and therefore minimum energy dissipation. At some intermediate velocity (time scale), there will be maximum dissipation, often with pronounced stick-slip.*

*This argument should be quite general and will apply at various length and time scales. McClelland's Tomlinson-type model (see page 406) may be analyzed in this way. In other cases the length scale may be determined by molecular or asperity dimensions, or may be macroscopic, as in the case of a squeaking door."*

Several other issues on energy dissipation and time and length scales are listed below:

- 1. What are the energy dissipation mechanisms in friction processes? some suggestions are: lattice excitations (phonons) and heat generation; Schallamach and Stoneley waves-generation and propagation; phase transformations (amorphization, glassification, solid-liquid, etc.); internal friction (e.g., in polymers)...**
- 2. At the atomic scale, can some of the vibration energy be used to overcome the potential barrier of the next friction event or is all of it dissipated into the bulk?**
- 3. Are there different characteristic length and time scales associated with static friction and kinetic friction?**
- 4. While time and length scales are certainly important, what other scales may be pertinent to friction processes, e.g., film thickness/contact length or molecular vibration period/sliding speed?**
- 5. Is energy dissipation a necessary consequence of discontinuous motion during friction?**

### **3.11 NEW WAYS OF PROBING FRICTION PROCESSES.**

Why do we know so little about friction processes? Stated simply by Tabor, "*we do not have a way of seeing what is actually taking place at the interface while sliding is taking place.*" Hence, one of the experimental difficulties in investigating friction is that friction occurs at a buried interface. A second difficulty is that friction events can take place rapidly: less than nanoseconds for atomic events and microseconds for micrometer-sized asperity events. New approaches are therefore needed to investigate, in real time, the prompt loss processes buried in the interface of a sliding junction.

Pethica, Tabor, Robbins and others have suggested several spectroscopies to investigate friction in the buried interface:

- 1. Optical spectroscopy** To gain access to the interface, make one sliding face sufficiently thin or "transparent" to the radiation. An example would be the real-time optical spectroscopies of elastohydrodynamic (EHD) fluids being performed at Shell Research Ltd. and at Imperial College (by Spikes et al, see end of part 4). The work partially bridges the gap between macroscopic and molecular processes in EHD lubrication.

Interference gives the film thickness and shape down to vanishing film thickness; the contact zone is scanned by a laser and the Raman shift is used to determine the pressure distribution in the film; then optical birefringence gives a measure of molecular orientation in the film during flow.

To resolve tribochemical processes at the picosecond time scale, time-resolved pulsed laser spectroscopy was recommended to probe the reactions, possibly in conjunction with pulse energy to trigger events. Non-linear spectroscopies of buried interfaces were also described by Dick Polizzotti.

2. Electromagnetic wave probes using e.g. microwaves.
3. Phonon spectroscopy. Perform experiments at liquid He temperatures, where phonons have longer lifetimes, and use high quality (low phonon scattering) substrates to transport the prompt phonons ballistically into a detector. Determine from the phonon energy spectrum whether the energy at the interface appears in vibrational, translational or rotational form.

Pethica then recommended measurements of friction at small time scales, since many chemical processes and the molecular dynamics simulations of these processes take place in a time domain below a nanosecond. But, he cautioned, it may be exceedingly difficult to resolve forces on a time scale less than 10  $\mu$ sec, because of the mechanics of an apparatus. To follow the mechanical response e.g. stress/strain behavior in real time, an apparatus would have to have a sub-nanosecond response time. However, calculations show that such small response times require that the mass of the apparatus be impractically small e.g. only a few atomic masses. Even present day atomic force measurements, coupled through a fine tip, find it hard to achieve a time constant less than 10  $\mu$ sec. As a result, the slower responding apparatus cannot see the true behavior, but only a response curve dictated by the time constant and load-line of the apparatus. Therefore, there is a great incentive to make these measurements **indirectly**, and the scientist who comes up with the method will certainly be breaking new ground. [Several "unorthodox" friction probes are mentioned at the end of part 4.]

Uzi Landman suggested an approach for determining the role of chemisorbed films in friction processes, an important issue to understanding boundary lubrication. The approach combines theoretical modeling of phonons interactions with states of chemisorbed molecules and experimental infrared spectroscopy that gives information on lifetimes and transitions for stretched or distorted molecules on a surface. If we know the molecular distortions produced by friction and the distorted states of the molecule, then we can calculate how energy is dissipated by friction and how dissociation occurs on the surface.

In addition, there are still many investigations that can be made using more conventional friction probes:

1. **Investigate complex motions (load-axis spin, periodic loading) using a traditional friction tester.**
2. **Proximal probes: friction at controlled microstrain, at ultralow loads, ...**

#### 4. Hierarchy of Friction Models.

#### 4.1 FRICTION OF MACHINES.

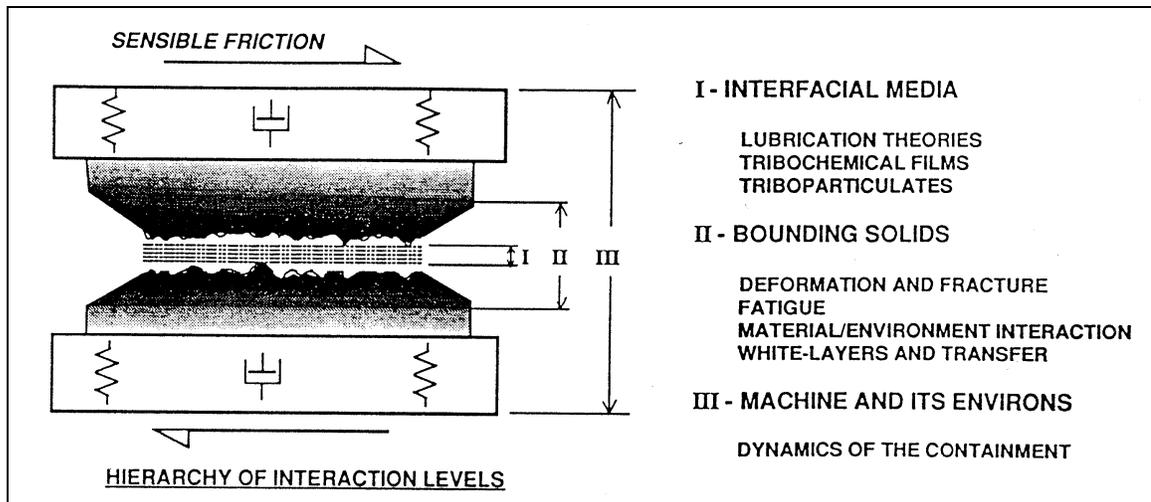


Fig. 1. Interaction levels in a friction machine.

Peter Blau pointed out that the entire tribosystem must be considered when modeling friction. He suggested<sup>1</sup> that we develop a hierarchical model based on where the shear forces are concentrated, from the interface out to the machine itself. Friction in a machine is often influenced by physical processes operating at more than one location and size scale in the tribosystem -- from the atoms and molecules interacting in the lubricant at the interface to the bolts holding the machine to the floor. Start with an interface between two moving surfaces and work outward (see Fig. 1). If the surfaces never make contact, we can probably model friction accurately knowing only the properties of the media (I) between the surfaces. If however there is solid-solid contact, the topography, chemistry and mechanical properties of the bounding solids and third body particulates must be considered. Finally, if shear stress cannot be accommodated within those layers, the shear forces will be transmitted beyond the near-surface zone (II) to the constraining machine and fixtures (III) and result, perhaps, in vibrations which may feedback from the machine to the interface.

#### 4.2 ATOMISTIC APPROACHES FOR ENGINEERING SOLUTIONS.

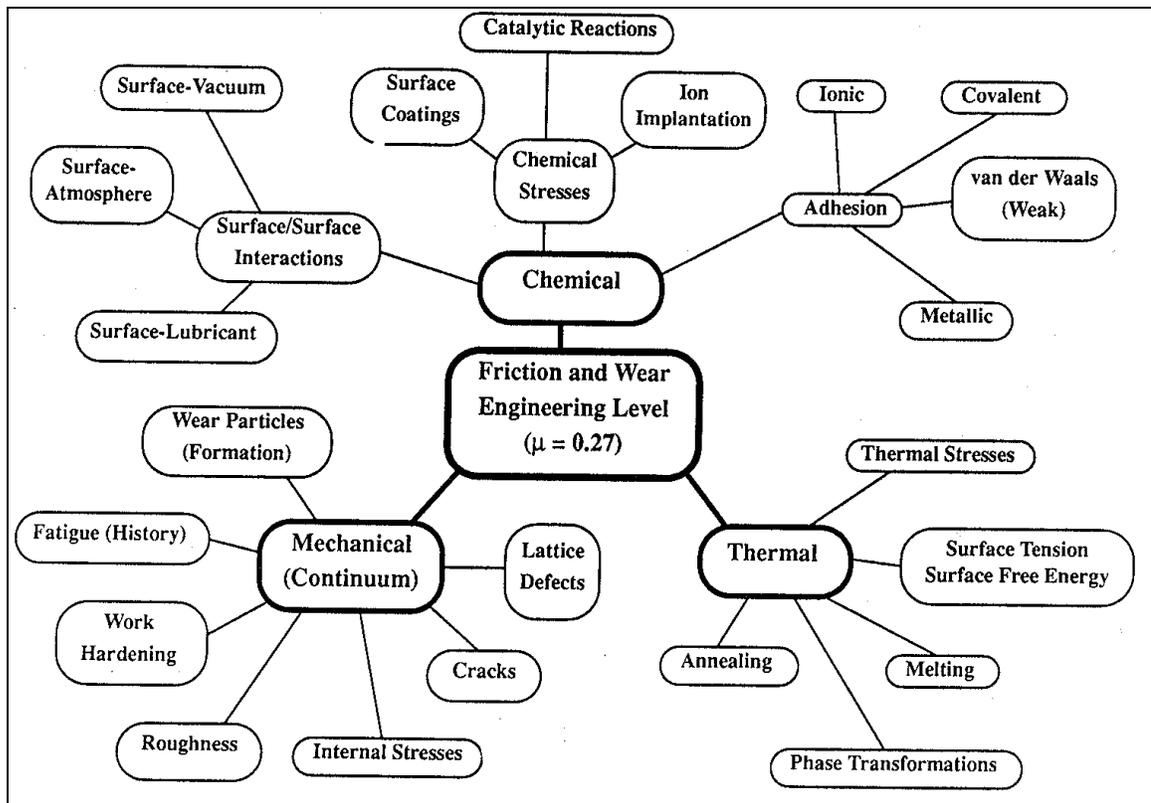


Fig. 2. Hierarchy of inputs required to calculate friction coefficient at an engineering level.

Much has been said at this Institute about atomistic experiments. Hence it would be valuable to consider how such experiments could be used in the reliable and efficient manufacturing of sensible products, i.e., how can atomistic methods make an impact at the engineering level? Jim Belak and his group of wandering scholars, including Kristian Glejbol, Dieter Klaffke and Arvin Savkoor, came up with a chart that symbolizes the transition from the atomistic approach to the engineering level (Fig. 2).

They started at the engineering level where it was agreed that a single number, namely the coefficient of friction, is adequate for designing a machine and assign it a reasonable friction coefficient: 0.27. The various levels of understanding needed to apply an atomistic approach are represented in the tree diagram. The Friction and Wear Engineering Level at the center of the tree is characterized by  $\mu = 0.27$  and represents the least amount of fundamental understanding. At the outermost branches, they put atomistic descriptions of the materials and their interactions, about which much is known, both experimentally and theoretically.

Although three areas of tribological phenomena (Thermal, Chemical and Mechanical) are represented, the mechanical branch will be discussed in order to illustrate the hierarchy of levels of understanding. Moving away from the center, one first encounters a continuum elastic/plastic description with a single yield criterion ( $Y = \text{constant}$ ). At the next level, there

are polycrystalline grains which can produce inhomogeneous and anisotropic behavior in a material. Then, within each grain, deformation is controlled by dislocations and other defects. These defects are ultimately controlled by the interatomic interactions at the outermost perimeter of the branches.

#### 4.3 THEORETICAL MODELS AND EXPERIMENTAL APPROACHES.

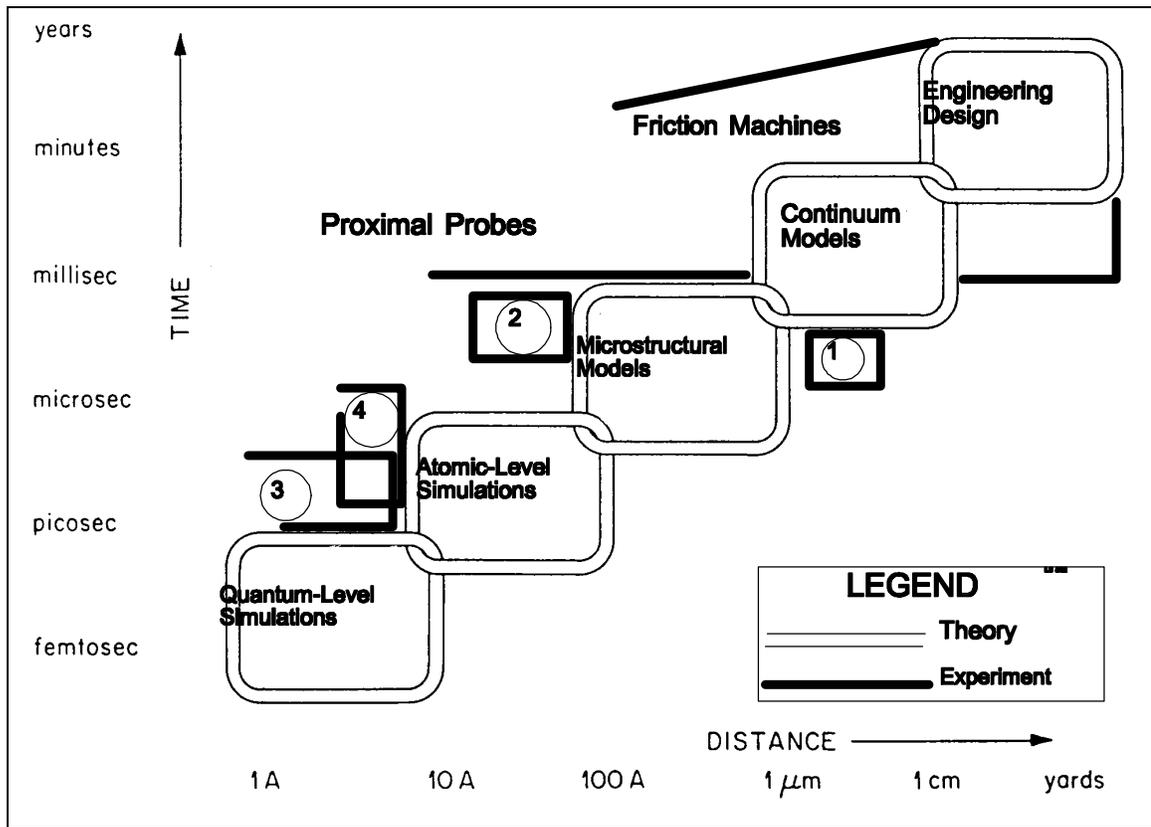


Fig. 3. Time and length scales of present-day models and experiments in Tribology.

How can we use the power of microscopic modeling to gain new insights into macroscopic friction processes and, ultimately, to solve technological problems? Bill Goddard<sup>2</sup> suggests that this can be done by progressing along the "chain-linked" ladder, illustrated in Fig. 3, from quantum-level studies to engineering design. His "hierarchy of modeling tribological behavior" unites atomistic models, which operate in very short length-time scales, with engineering models, which describe tribological behavior in length-time scales perceivable by more traditional measuring equipment. This approach "...allows consideration of larger systems with longer time scales, albeit with a loss of detailed atomic-level information. At each level, the precise parameters (including chemistry and thermochemistry) of the deeper level get lumped into those of the next. The overlap between each level is used to establish these connections. This hierarchy allows motion up and down as new experiments and theory lead

to new understanding of the higher levels, and new problems demand new information from the lower levels."

But where are the experimental approaches for investigating the "lower (short scale) levels?" As illustrated in Fig. 3, most "friction machines," including the proximal probe devices, are operated at long time scales. An abbreviated search of the recent literature by one of the editors, Irwin Singer, found only three "tribology" tests and a fourth proximal probe method that come close to investigating friction behavior at short time and length scales:

1. Bair et al.<sup>3</sup> have used fast IR detectors to measure flash temperatures during high speed frictional contacts of asperities of length 10  $\mu\text{m}$  and greater, with time resolution of about 20  $\mu\text{sec}$ .
2. Spikes et al. have developed *real time* optical techniques for investigating the physical behavior of EHL films down to 5 nm thick<sup>4</sup> and chemical processes occurring in contacts 10  $\mu\text{m}$  wide by 80 nm thick<sup>5</sup>.
3. Krim et al.<sup>6,7</sup> have used a quartz crystal microbalance to measure the sliding friction of molecularly thin films (both solid and liquid) condensed onto surfaces, probing atomic vibrations amplitudes between 0.1 to 10 nm and time scales from  $10^{-12}$  to  $10^{-8}$  sec.
4. Hamers and Markert<sup>8</sup> have shown that STM images are sensitive to the recombination of photo-excited carriers whose lifetimes are in the picosecond range.

Clearly, innovative experimental approaches for measuring friction processes at short and intermediate time-length scales (like those outline by Pethica earlier) are needed to assist the modelers who are already there.

Finally, there is another time scale to consider, and that is "the question of time to translating this (fundamental) knowledge into engineering practice." Duncan Dowson tells us how this can be accomplished in the next and final part of the Epilogue.

## 5. Commentary by Duncan Dowson.

*"I think that before making a few observations on the meeting itself, I would like to take this personal opportunity to thank the organizers for providing me with the opportunity to attend. It has been a remarkable experience for me. I would echo David Tabor's comment that this has been the most valuable meeting in the subject of friction that I have ever attended.*

*We have heard quite a lot about the subject of friction from the atomic scale up to the macroscopic scale. I suppose that we are all reflecting this morning on the issues we clarified and the points that we will take back in our minds to our laboratories or to our computers and to determine how the information that we have received will affect the direction of our future research. The answer will be different depending upon where you come from, what your background is, whether you are a physicist concerned with the atomic scale or whether you are an engineer and trying to design the next generation of jet engines.*

*In some ways we could take a pessimistic view of the outcome this fortnight. What have we heard that we did know about before we came to this meeting? I think we should all ask ourselves that question. If the answer in some cases is negative, I think we should reflect further on the point already made this morning that the new bits of information that have come in strengthen our confidence and understanding of the subject as a whole. A lot of loose ends have*

been brought together and clarified very effectively by the participants, the speakers and the discussers, and I have found that very valuable.

There has, of course been a great deal of discussion on size effects, a subject of great importance to both solids and fluids. In general terms, we have been talking about length scales -- angstroms and nanometers up to micrometers. Little has been said this morning about fluids and indeed little has been said at the meeting about friction associated with fluids. But we have heard a few very revealing observations about the behavior of fluids adjacent to solid boundaries. In a sense, as I mentioned earlier, I take great comfort in the fact that these remarkable changes in fluid behavior do seem to be limited to very small distances from the solid boundary. And since most engineers are talking about effective fluid films which are a few orders of magnitude bigger than those distances, you might argue that the boundary effects as such may not be very meaningful. While they certainly provide details of boundary behavior under no-slip condition, whether dry or not, and indicate unusual viscosities of molecularly-thin layers adjacent to the boundary, do they really affect the outcome of our overall calculation? The answer is no as far as I can see. We can confidently apply continuum mechanics at the present scale.

But the present scales **are** related to effective lubricant film thicknesses. Jim Greenwood and others have demonstrated that we are now starting to talk about nominally smooth surfaces in which the effective film thickness is on the order of the roughness of protuberances on engineered surfaces. Will it be difficult to separate smooth surfaces by coherent films of a lubricant? These surfaces are going to come very close together and touch on asperities either in steady state motion or certainly while starting and stopping machinery. And so we are concerned with the effects we have heard about here, both in relation to solid friction at such interactions and of course in relation to the effects on the lubricant properties and behavior within two molecular layers from the surface.

There are other aspects of scale I think we should reflect on. One is the question of time in terms of translating this knowledge into engineering practice. Let me assume that you have heard something during the last fortnight that might have an impact on the design and operation of machinery. You may be impatient to wait to see how long it takes for that to show itself in practice. The time scales are generally **enormous**. We are not talking about next month or next year. There are engineers out there who are already designing the motorcar engines for the year 2000 and jet engines for the next century. Before there can be a translation of some of the concepts, and that assumes you have new concepts, I think you should take note that it is going to be about a decade if not a generation before that impact will be seen fully in engineering. This makes it all the more important for meetings like this to bring together physicists, chemists and engineers, so that the engineers can absorb by osmosis the concepts that you are revealing. Don't shut us out just because it will take us 10 years or 20 years. It is important that these ideas feed into our consciousness so that we apply them sensibly in future developments.

Little has been said about the subject of wear at the meeting. Perhaps I was slightly surprised that there was not greater emphasis and greater attention paid to the results of friction, and whether friction processes are intimately related to production of wear particles. What is the morphology of the wear particles? Is there anything to be learned from them that we could feed back into the understanding of the friction process? For example, there are many revealing studies in the field of wear about the size and the nature of wear particles. About a year ago, I heard Professor Kato from Japan give an illuminating talk. He would have us believe that as

*I walked across this wooden floor this morning, with every step I took, between 1000 and 10000 wear particles were produced between my shoe and the floor. Maybe the size and the shape of the wear particles have something to say about the processes involved, the interactions of the solids associated with friction and wear. Perhaps we should have devoted a little more time to that interface than we had time to do. (Maybe Irwin you should start thinking now about the subject for your next ASI.)*

*In concluding, I would like to say that there is also another aspect that I think is important. As I listen to physicists and chemists discussing in intimate detail the atomic interactions, I think at times there tends to be a feeling of competition between science and engineering. That I think is totally wrong and misleading. Certainly from the engineers' point of view, in my experience, we love the opportunity to try to hang on for those physical concepts, to the understanding of basic interactions in solids and in liquids and all phases of matter. We may not fully understand all of them, but it is most important that we try to keep the communication as strong as possible. I hope equally that scientists will not be too frustrated by the fact we take 10 to 20 years to incorporate their bright new ideas in designing better skis or whatever it might be. Let us perpetuate this interaction between groups of people who all have one objective and that is to understand the laws of physics in order to apply them as effectively as we possibly can for the good of society through the manufacture of reliable, efficient and sensible products."*

## **REFERENCES**

1. P.J. Blau, STLE Trib. Trans., 34 (1991) 335.
2. "Tribology of Ceramics" National Materials Advisory Board Report 435, National Academy Press, 1988, Washington DC, p. 88.
3. S. Bair, I. Green and B. Bhushan, J. Tribology 113 (1991) 547.
4. G.J. Johnston, R. Wayte and H.A. Spikes, Tribology Transactions, 34 (1991) 187.
5. P.M. Cann and H.A. Spikes, Tribology Transactions, 34 (1991) 248.
6. J. Krim and A. Widom, Phys. Rev., B38 (1988) 12184.
7. J. Krim, D.H. Solina and R. Chiarello, Phys. Rev. Lett., 66 (1991) 181.
8. R.J. Hamers and K. Markert, Phys. Rev. Lett., 64 (1990) 1051.

Irwin Singer and Hubert Pollock  
May 1992