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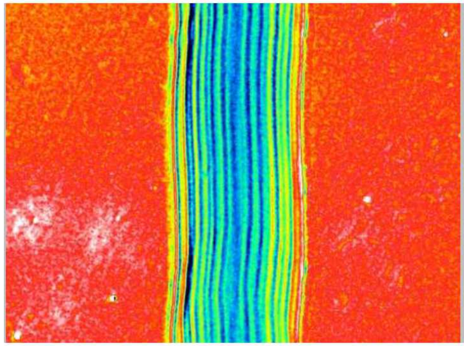

Development of Friction Models and Solid Lubricant Formulations for Extreme Environments

M.T. Dugger[1], J. Curry[1], A. Hinkle[1], N. Argibay[1], M.E. Chandross[1], B.L. Nation[1], and A. Korenyi-Both[2]
[1]Sandia National Laboratories, Albuquerque, NM; [2]Tribologix Inc, Golden, CO

MTSE Department Seminar, University of North Texas
16 November 2018

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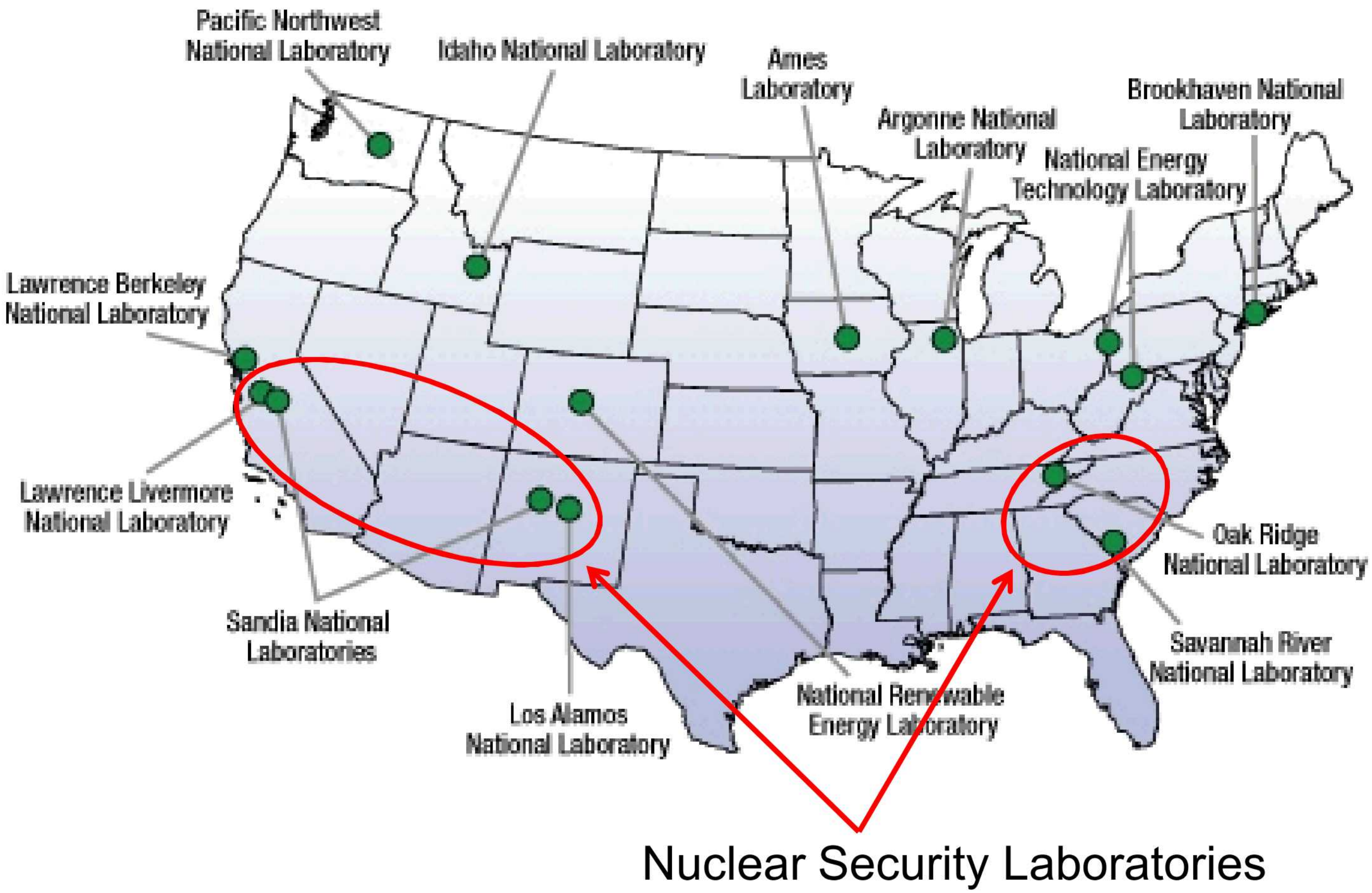
Sandia National Laboratories

A U.S. Department of Energy multiprogram laboratory, with responsibility for the safety, security and reliability of the nation's nuclear deterrent.



Sandia's Sites

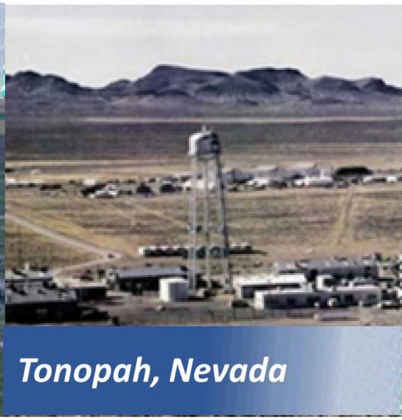

Albuquerque, New Mexico


Livermore, California


Kauai, Hawaii

Waste Isolation Pilot Plant, Carlsbad, New Mexico


Pantex Plant, Amarillo, Texas


Tonopah, Nevada

Sandia's National Security Mission

Nuclear Weapons

- ensure a safe, secure and reliable nuclear deterrent

Defense Systems & Assessments

- maintain U.S. military weapon systems superiority

Energy & Climate

- ensure clean, abundant and affordable energy and water

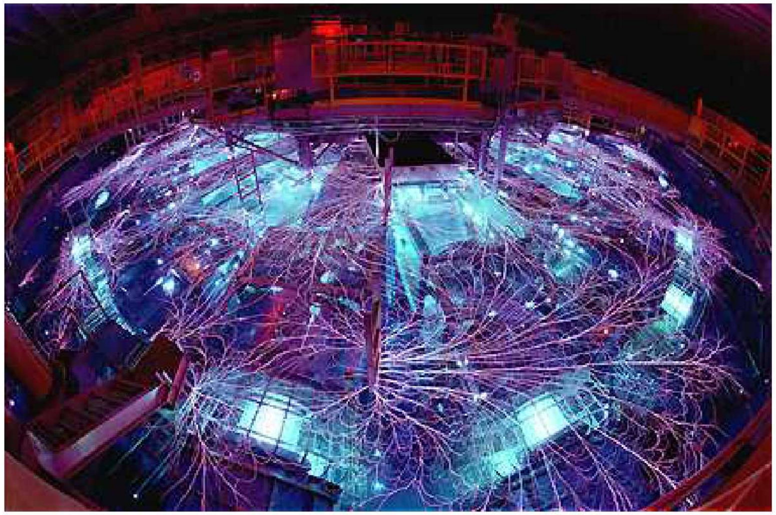
International, Homeland & Nuclear Security

- protect against terrorism through advanced technology

Science and Engineering Foundations

- R&D to support all national security missions

Sandia National Labs Programs



pulsed power and fusion research

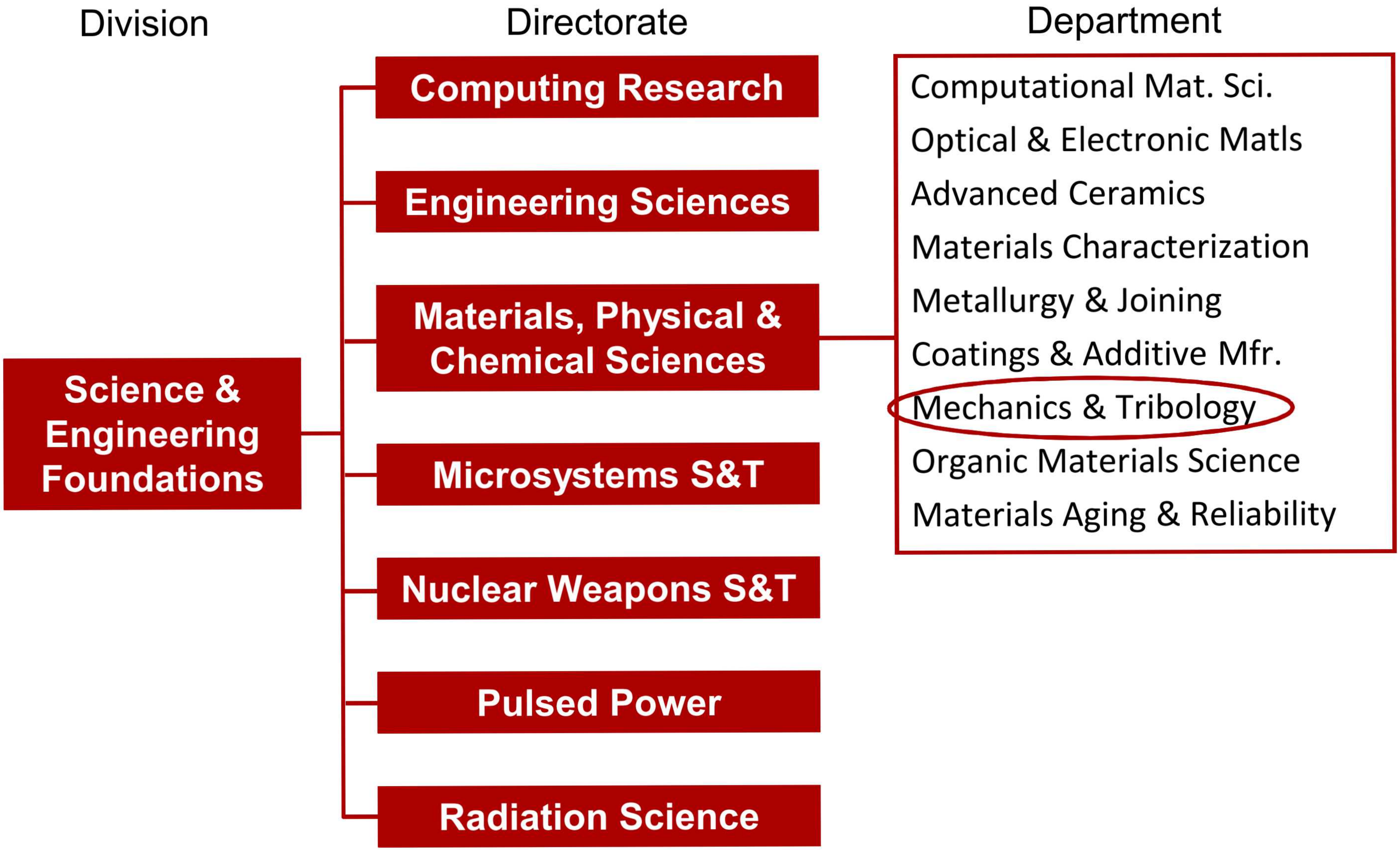
DoD systems design and testing

renewable energy

Plus a host of other fundamental to applied sciences

- physical and chemical sciences
- high performance computing
- radar and guidance
- satellite systems
- geological sciences
- biotechnology…etc.

Sandia's R&D Organization



2019 Tribology Portfolio

Research

- High J Electrical Contacts
- Solid Lubricant Aging and Performance
- Mechanical Stability of Nanocrystalline Metals
- Fluid Degradation at Sliding Surfaces

Work for Other Federal Agencies

- Reliability of Electromagnetic Launch System Components

Engineering Support

- Impact Resistant Hard Coatings
- High Current Electrical Contacts
- Tribological Coatings for Brayton Cycle Components
- Environmentally Robust Solid Lubricants for Miniature Mechanisms
- Damping Fluids for Accelerometers
- Reliability of Electromechanical Device Electrical Contacts

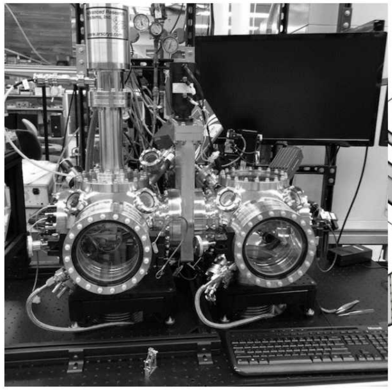
Tribology Team

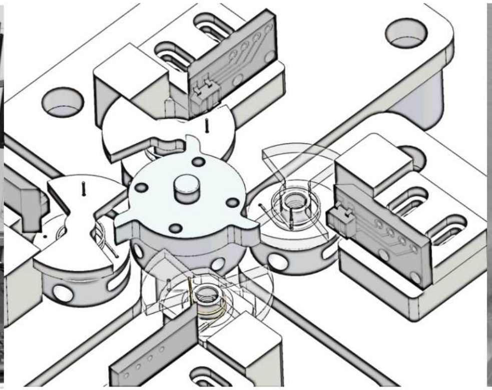
3 Technical Staff

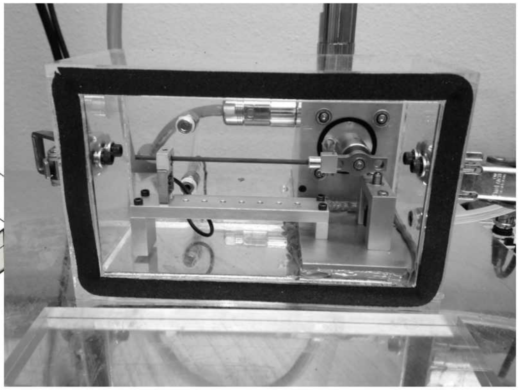
3 Technologists

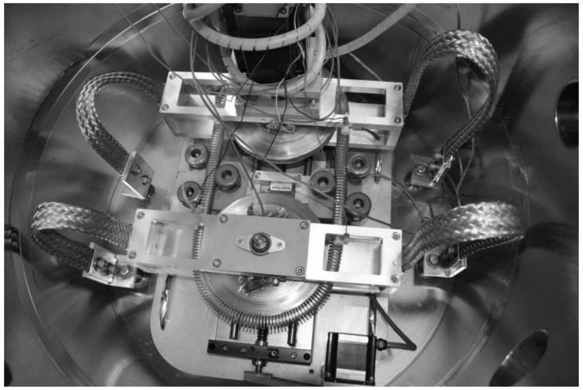
1 Post Doc

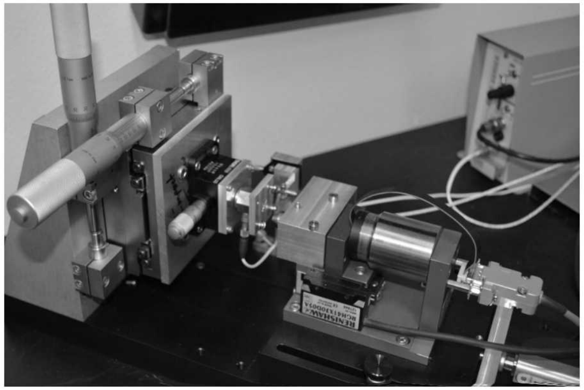
1 Student Intern

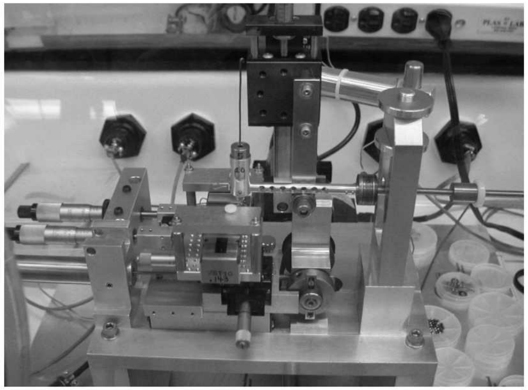












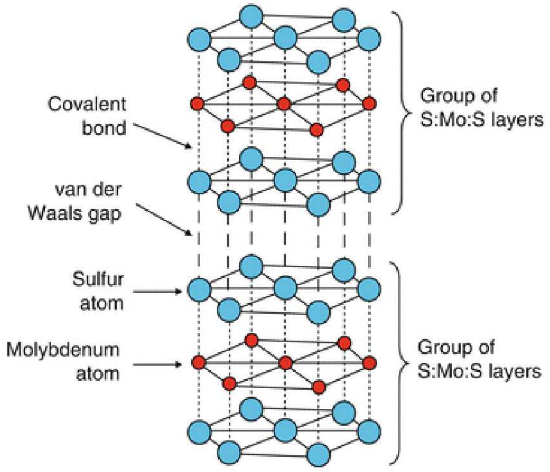
Liquid vs Solid Lubricants

Liquids

- prevent surface contact at sufficient speed (hydrodynamic)
- replenishable at real contact area
- provide some vibration damping
- remove heat and wear debris from contact

Solids

- wide operating temperature range
- friction less sensitive than liquids to shear rate or contact pressure
- generally not replenishable
- most exhibit sensitivity of friction coefficient to operating atmosphere



- Lamellar solids such as MoS_2 have weak bonding between layers, creating a low shear direction
- Shear causes orientation of the low shear plane parallel to motion
- Forms a “transfer film” on the mating body
- Sites at edges of crystallites are particularly reactive (oxidation, adsorption)

Extreme Environments

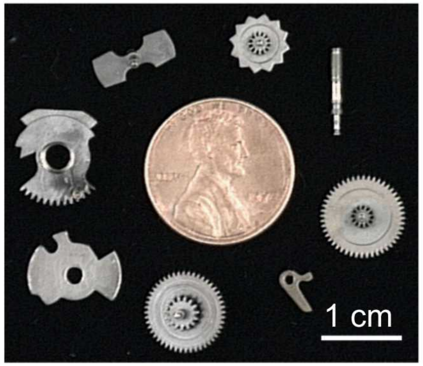
Sandia National Laboratories

Space:

- operate in vacuum (+atomic oxygen in low earth orbit)
- store months – years before use; generally non-serviceable
- operating temperatures from 50 – 300K, depending on location
- large investments of time and money

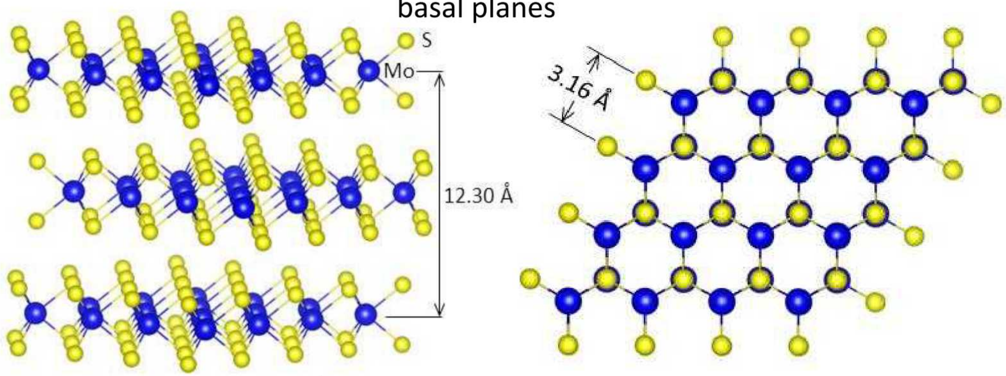
Precision Mechanisms:

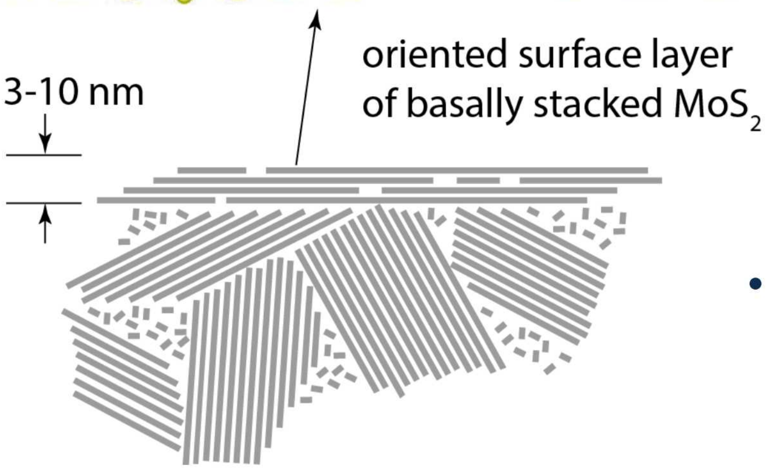
- inert gas near P_{atm}, trace O_2, H_2O, outgassing species
- store for *<u>decades</u>*; non-serviceable
- operating temperatures 200 – 350K
- large investments of time and money
- consequences (political, societal) of failure are unacceptable



MoS_2 Lubrication Mechanism

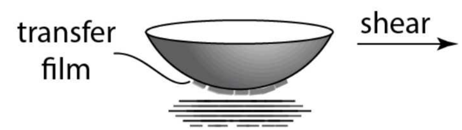
molybdenum disulfide
μ = 0.02 – 0.06 (inert gas/vacuum)
μ = 0.15 – 0.25 (humid air)



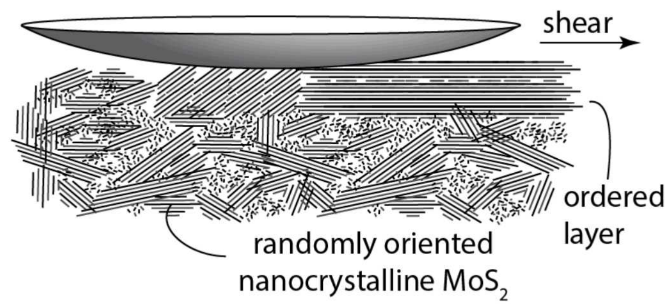


Run-In Processes:

1) Transfer Film Formation



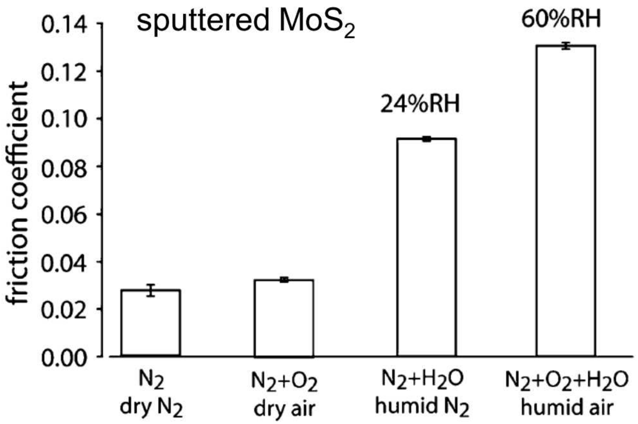
2) Shear-induced re-orientation and coalescence



- The *shear accommodation mechanism* of these materials is inter-lamellar sliding

Sliding occurs between relatively weakly bonded basal planes in MoS_2 and related lamellar solids

Effects of MoS_2 Aging

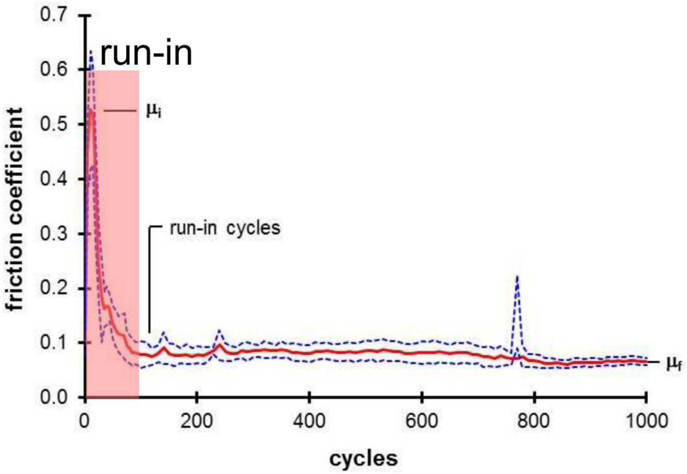


- Steady-state friction coefficient at 30°C of sputtered MoS_2
  - significant friction increase with water vapor increase in the environment
  - far less sensitive to oxygen

H. Khare and D. Burris, Tribology Letters **53** (2014) p.329-336

- Surface oxidation can dramatically increase the initial friction coefficient
  - atomic oxygen exposure resulted in oxidation of the top 100 nm of film

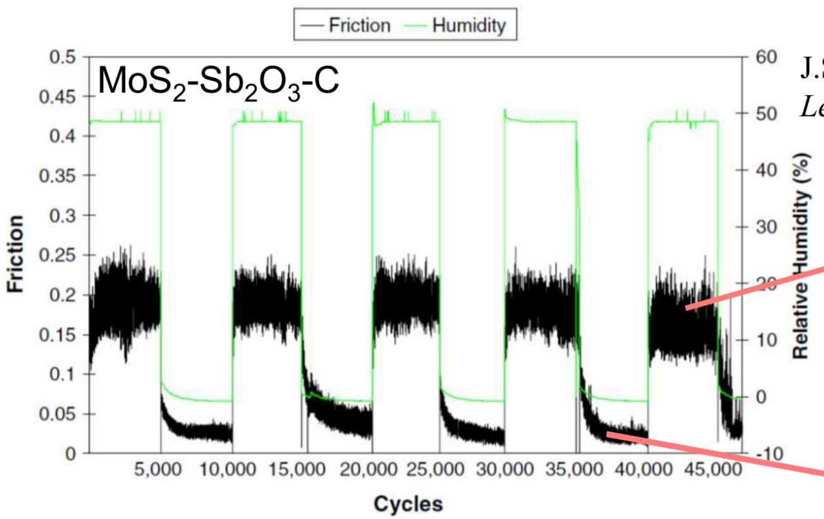
M.T. Dugger, T.W. Scharf and S.V. Prasad, *Adv. Mat. and Processes* **172** (2014) p.35-38



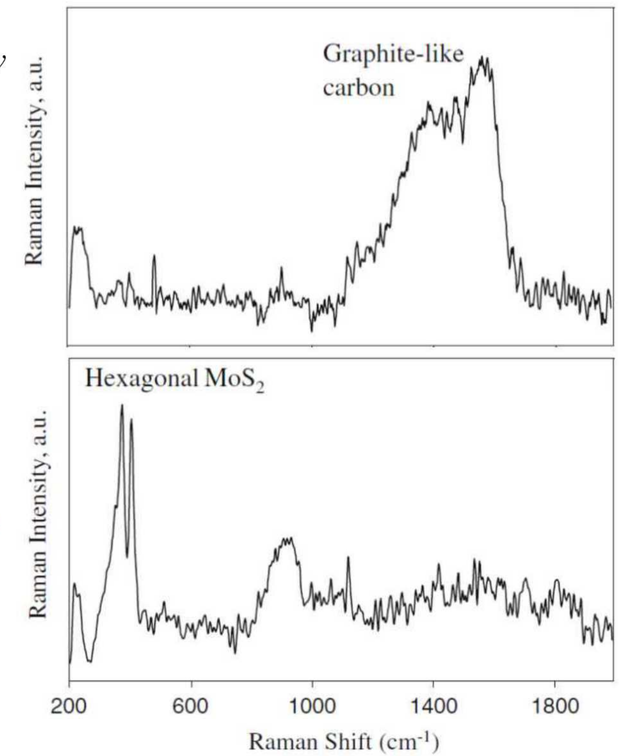
Some precision mechanisms' entire service life is spent in run-in

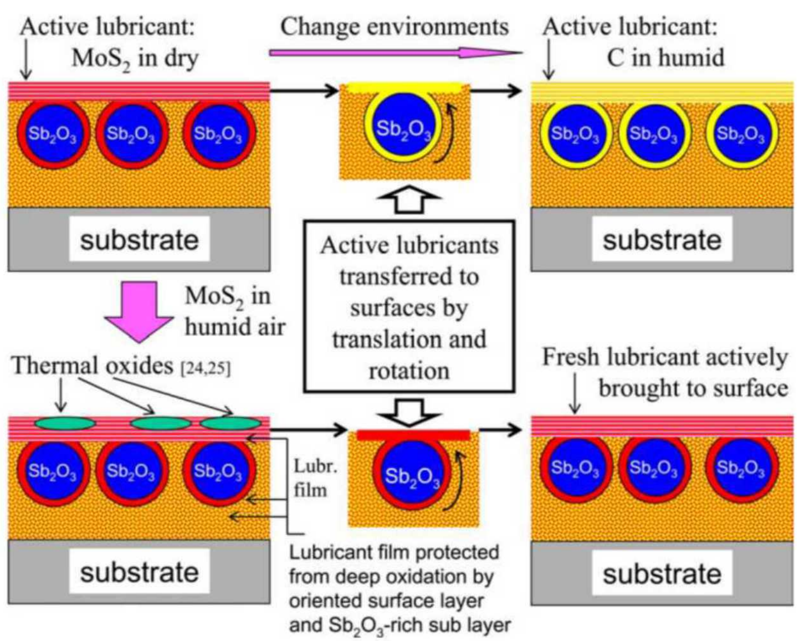
We desire a low and consistent friction response after years of dormancy

MoS_2 Composites: Burnished Films



J.S. Zabinski et al, *Tribology Letters* **23** (2006) 155





Phenomenological explanation:

- *oriented, low shear, non-reactive surface adaptively created by sliding*
- *sequester a reservoir of solid lubricant below the reinforcement phase*

Legacy Solid Lubricants and Processes

Resin-bonded films

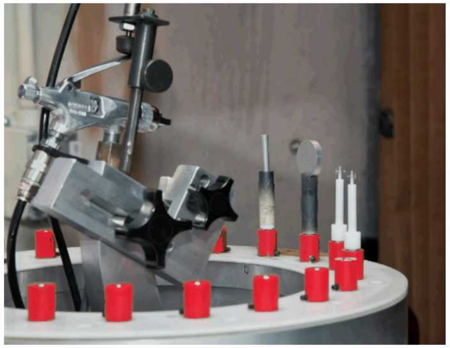
- blast surface with Al_2O_3
- steps: clean, mask, spray, cure, burnish, clean
- few parts at a time
- highly volatile organic solvents; carcinogens
- 2.5 μm *minimum* thickness

N_2 Sprayed MoS_2

- steps: clean, mask, spray, clean
- one to few parts at a time
- 200 nm *maximum* thickness

Mechanically Impinged MoS_2

- steps: clean, mask, tumble, clean
- batch process
- ~500 nm thickness (highly variable)
- can change part geometry

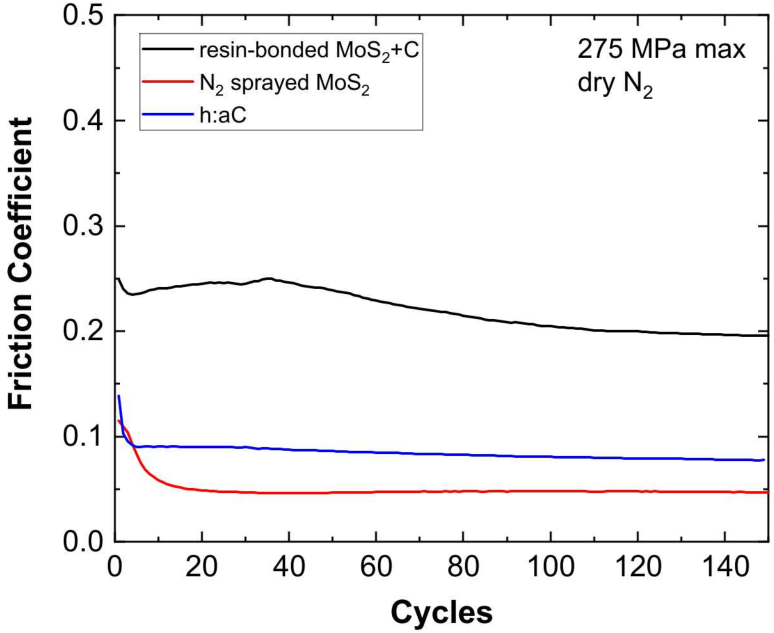


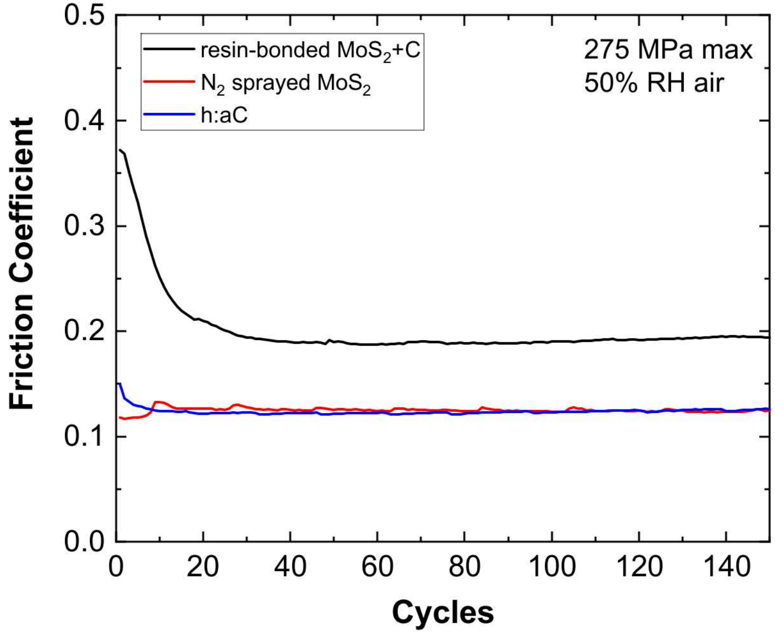
Spray MoS_2 + C with polymer binder

MoS_2 powder sprayed with N_2

Parts tumbled in drum with MoS_2

Friction Response of Legacy Materials





Resin-bonded MoS_2+C

- higher SS friction than impingement or DLC; process not suitable for high precision parts

N_2 Sprayed MoS_2

- environmentally sensitive; limited thickness yields low wear life; oxidizes in storage

DLC

- lower friction coefficient in all atmospheres is desirable
- brittle, subject to delamination in some impact environments
- can exhibit fluctuations in friction response, likely due to transfer film dynamics

Mitigation of MoS_2 Environmental Effects

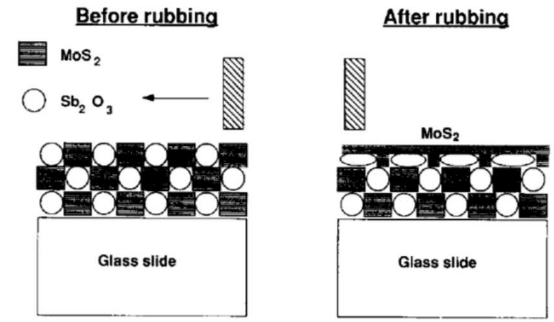
Sandia National Laboratories

Strategies

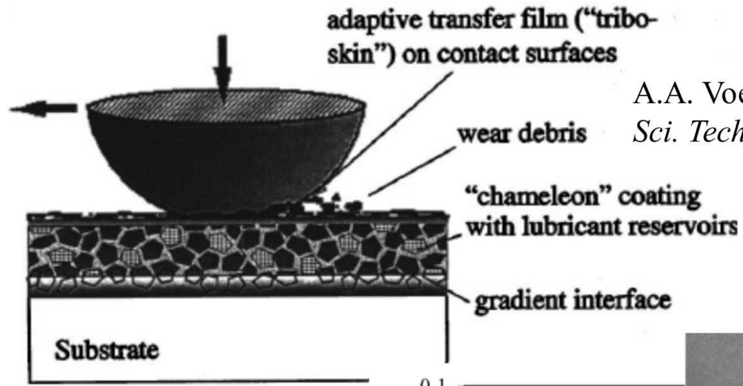
- dopants (Ni, Ti, Au, …)
- compositing - multilayers, multiple phases (Sb_2O_3, Ni, AuPd, …)
- ion bombardment during growth

Proposed Mechanisms

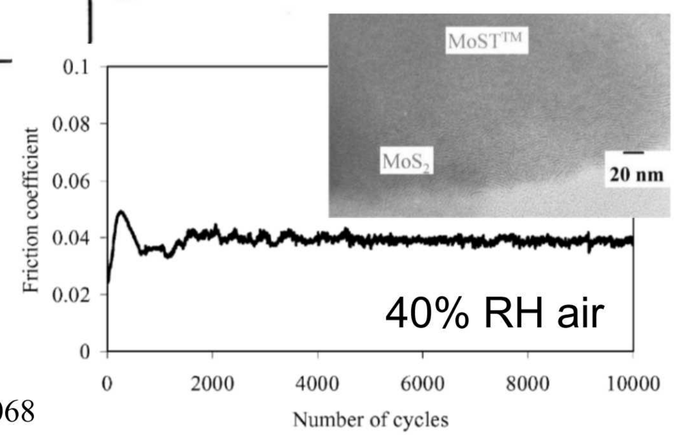
- densification
- increased hardness
- preferential orientation
- sacrificial oxidation of dopants
- passivation of MoS_2 edge sites
- crack arresting



J.S. Zabinski et al, *Wear* **165** (1993) 103



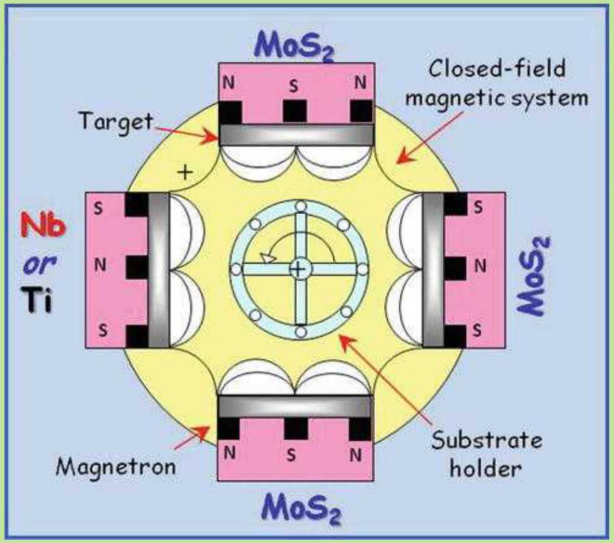
A.A. Voevodin et al, *J. Vac. Sci. Tech.* **A20** (2002) 1434



D.G. Teer, *Wear* **251** (2001) 1068

MoS_2 Composites: Sputtered Films

- Physical Vapor Deposition (PVD) is compatible with precise, small, complex parts, and facilitates quantitative quality assurance monitoring

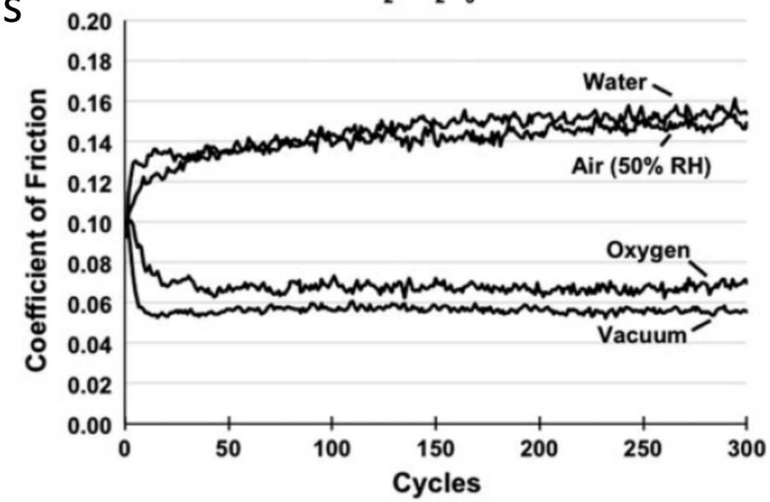


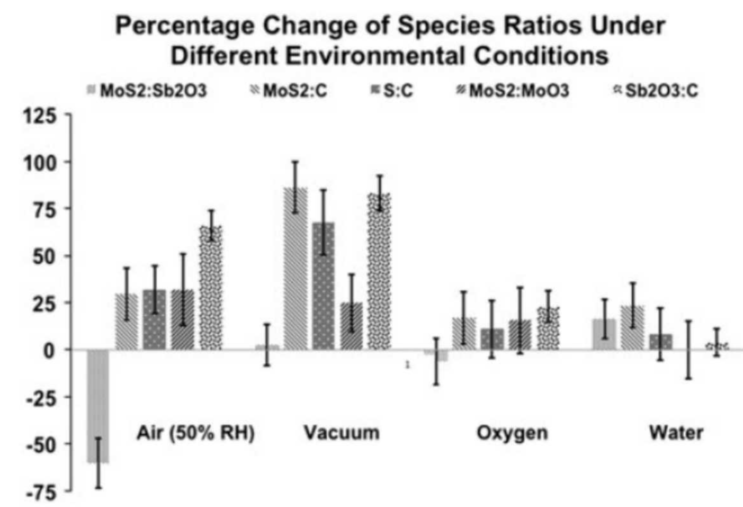
magnetron sputtering of MoS_2 composites

I. Efeoglu, in *Encyclopedia of Tribology*, Q. Wang and Y.-W. Chung, eds. (2013) Springer, p. 3233-3252

G. Dudder et al, *Tribology Letters* **42** (2011) 203

MoS_2-Sb_2O_3-C



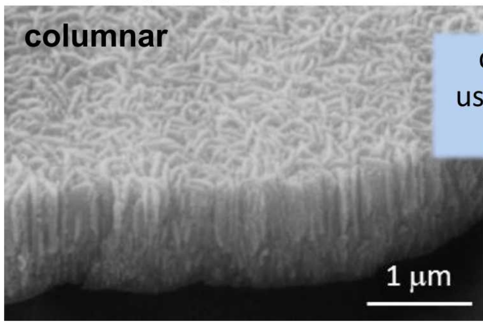


Examine variants of sputtered MoS_2-Sb_2O_3 and MoS_2-Sb_2O_3-C films for precision mechanisms

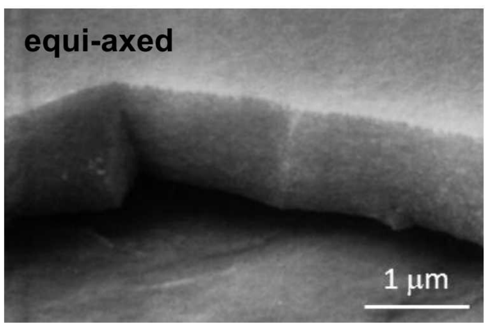
Sputtered Composites Facilitate Control of Film Structure and Chemistry

- Tailored film structures improve performance in a range of atmospheres
- Wear rates are improved by densification and inclusion of hard, load-supporting phases

M.R. Hilton, et al., *Surf. Coatings Tech.* **53** (1992) p.13-23

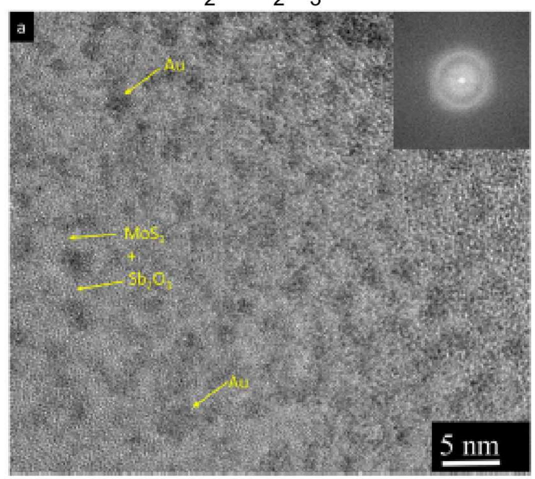


densification using deposition parameters

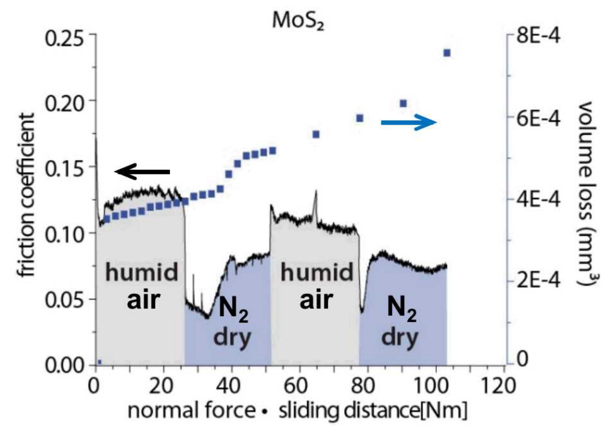


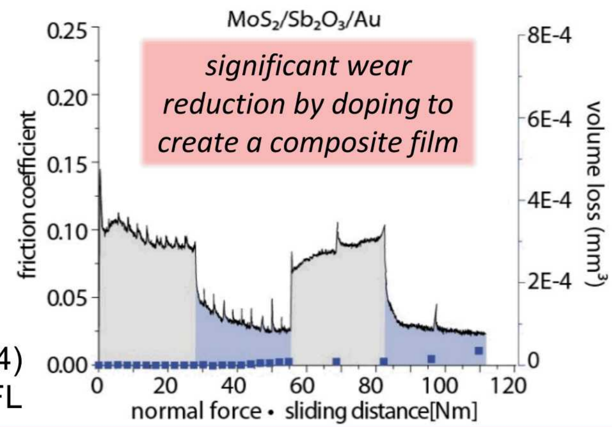
densification through composition control

$MoS_2+Sb_2O_3+Au$



H. Singh et al., *Surf. Coating Tech.* **284** (2015) p. 281-289





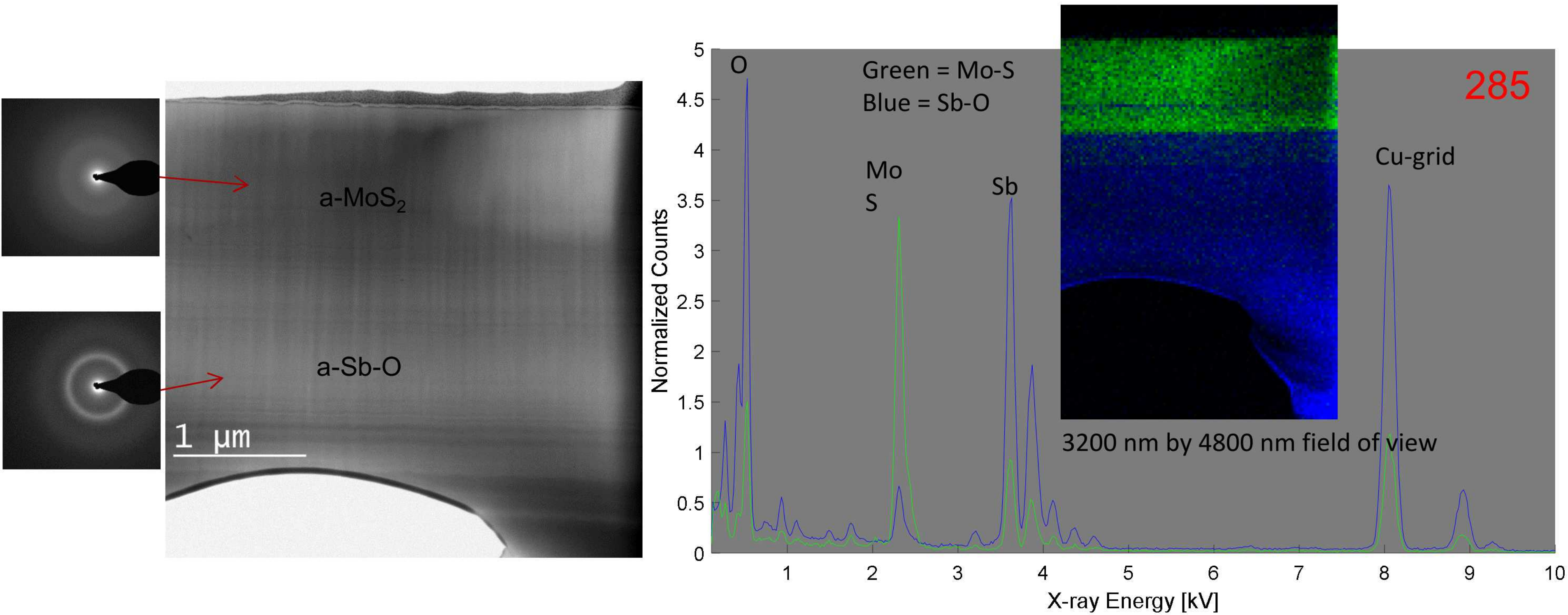
significant wear reduction by doping to create a composite film

R.S. Colbert, *Ph.D. Dissertation* (2014) U. of Florida, Gainesville, FL

Film Compositions Investigated

- MoS2-Sb2O3, I (285)
- MoS2-Sb2O3, II (2032)
- MoS2-Sb2O3-C, Bias
- MoS2-Sb2O3-C, Bias II
- MoS2-Sb2O3-C, Bias III
- MoS2-Sb2O3-C, Ion Assist
- MoS2-Sb2O3-C, Ion Assist II (282)
- MoS2-Sb2O3-C, Ti Bond (275)

RF magnetron sputtering



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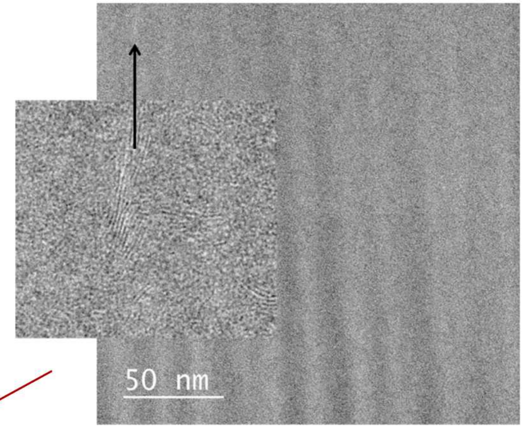
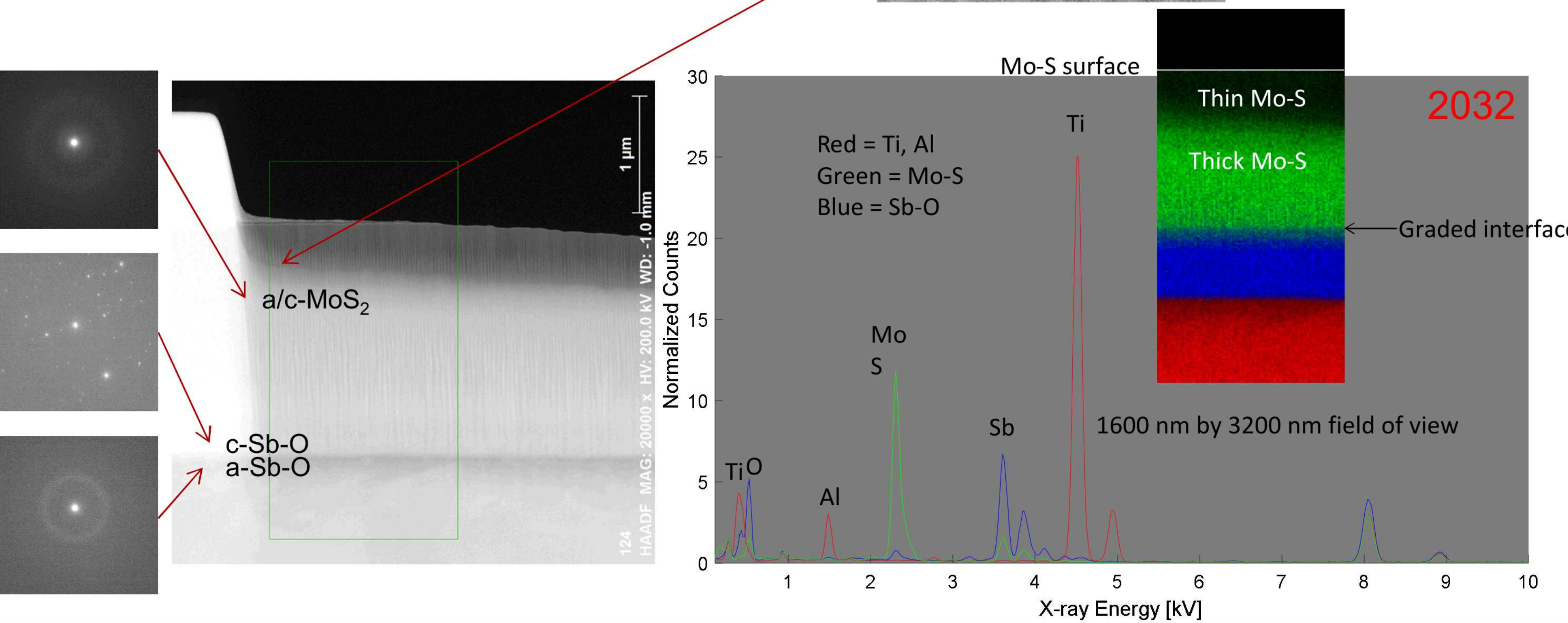


isolated local crystalline regions in amorphous matrix



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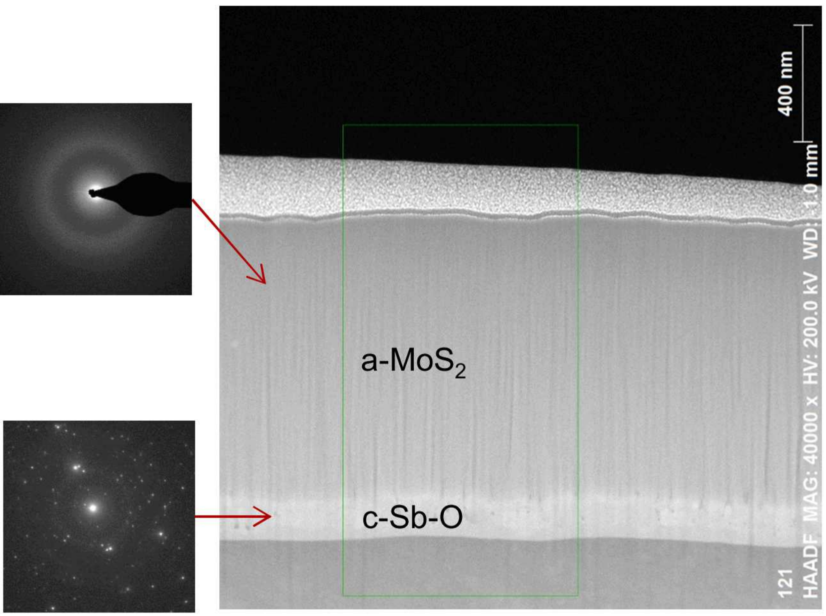

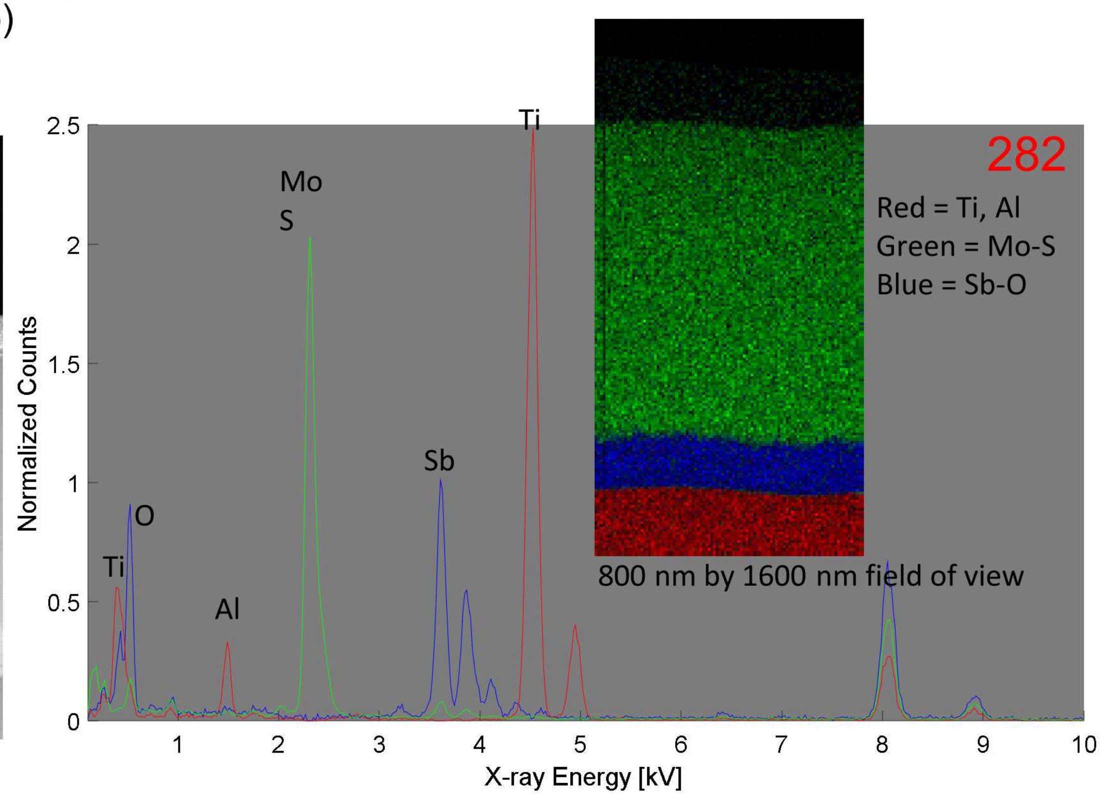

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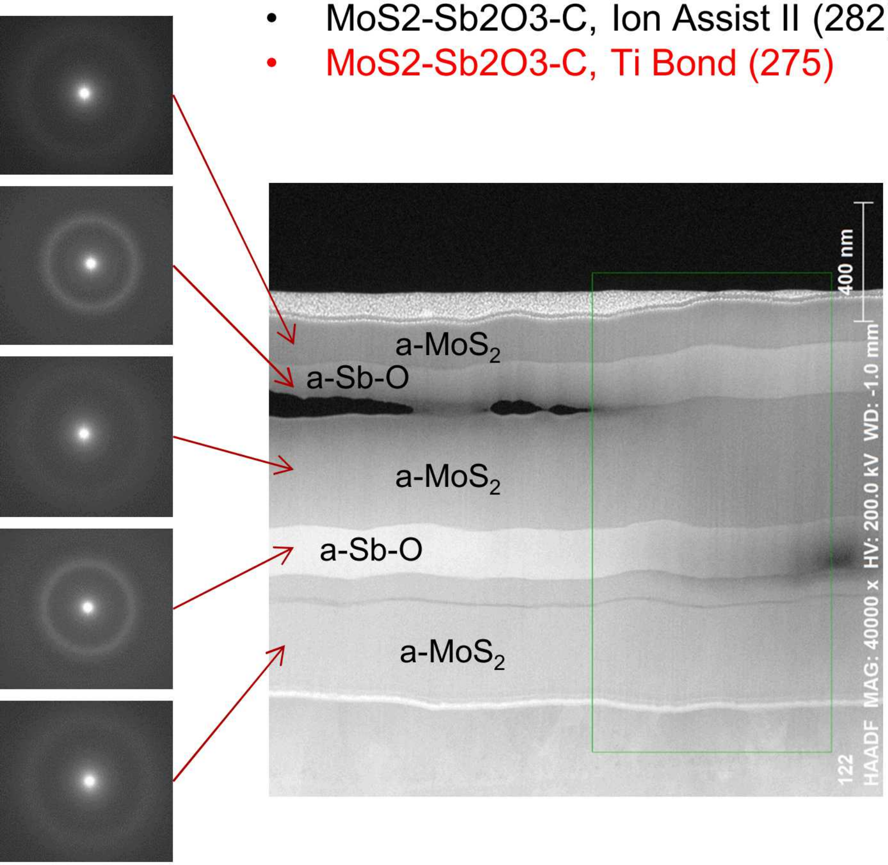
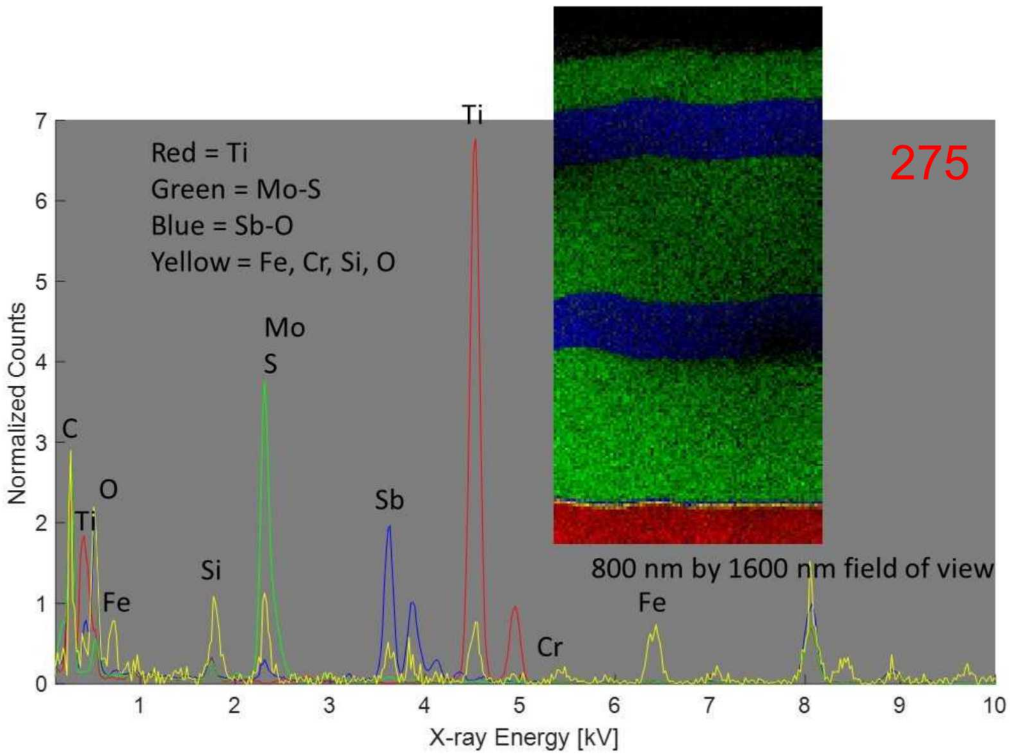
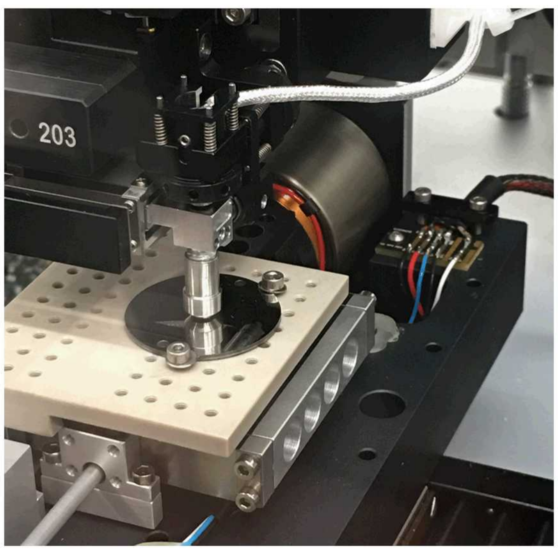
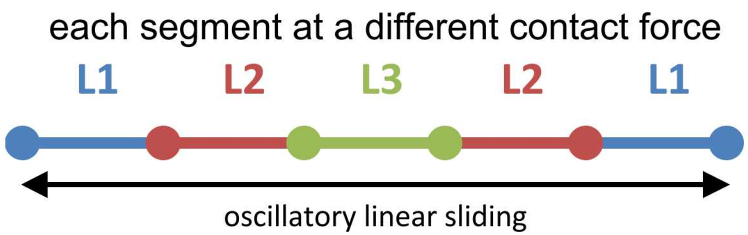




Friction Measurements: "Stripe" Tests

Load, mN	Max Pressure, MPa	Track Length, mm	Test Sequence	Cycles	Total Distance, mm
21	275	5	L1	300	1500
149	530	3	L2	500	3000
484	785	1	L3	1500	4500

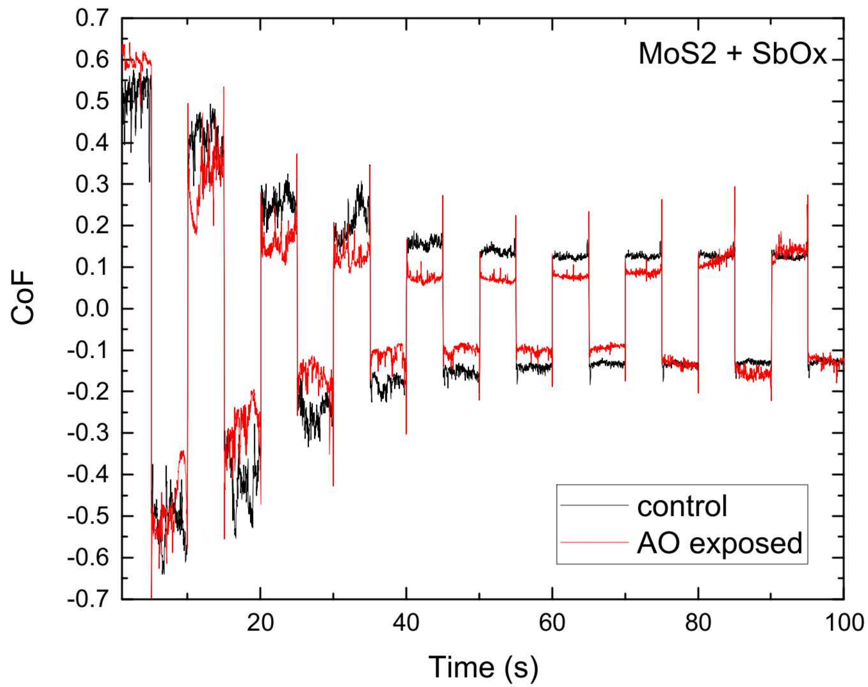




Test parameters:
- 440C ball, 3.2 mm diameter
- 1 mm/s sliding speed
- Controlled atmospheres:
  - dry N_2 (<10 ppm O_2, <50 ppm H_2O)
  - 50% RH air

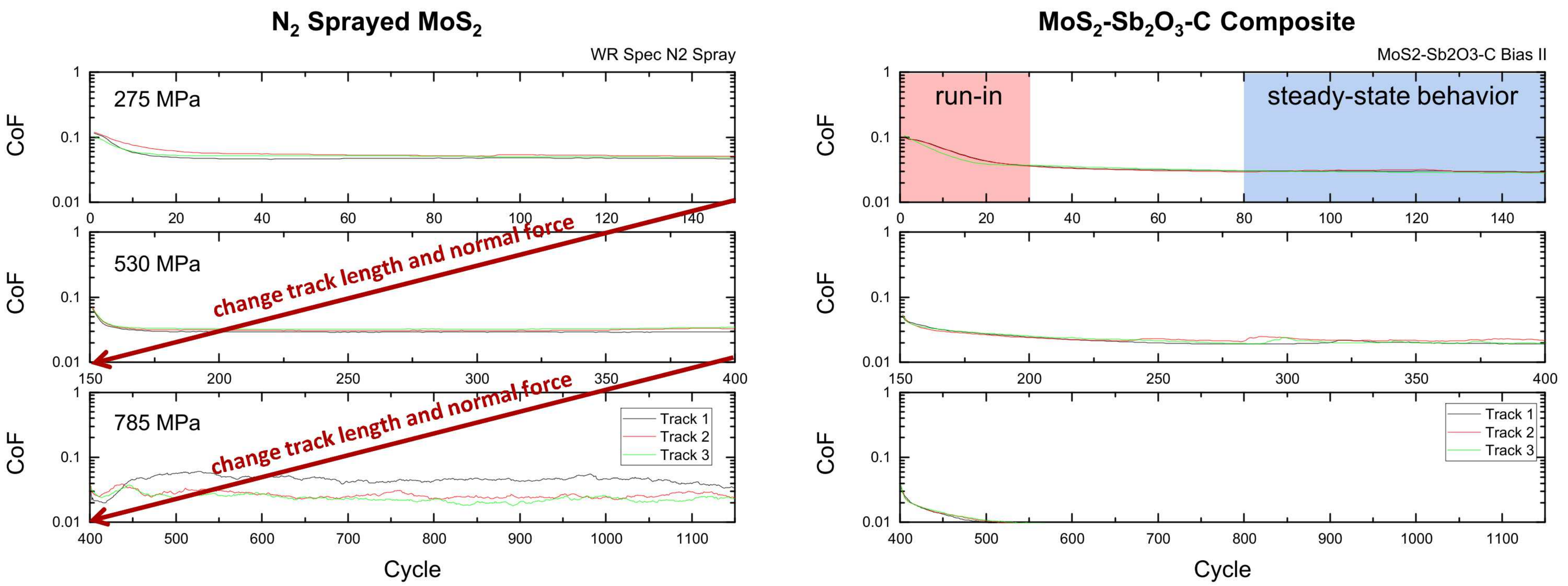
Permits performance assessment over a range of contact pressures

Raw Friction Data and Processing



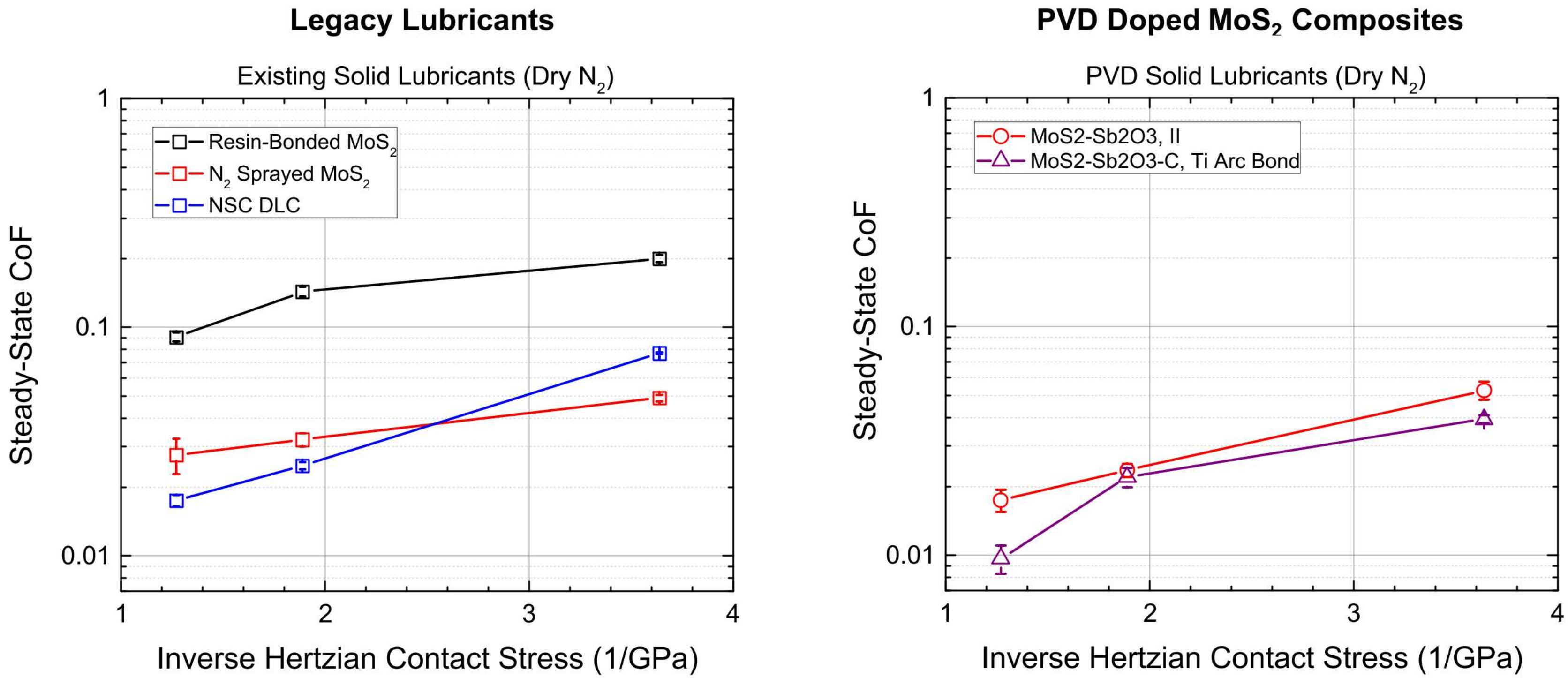
- Acquisition rate allows capture of run-in and friction variation within a single sliding cycle
- Bidirectional tests permit accurate measurement without drift
  - mask turnaround points and average adjacent bidirectional segments to get offset
  - difference between adjacent bidirectional segment averages is 2x friction coefficient

Friction Coefficient Traces from "Stripe" Tests



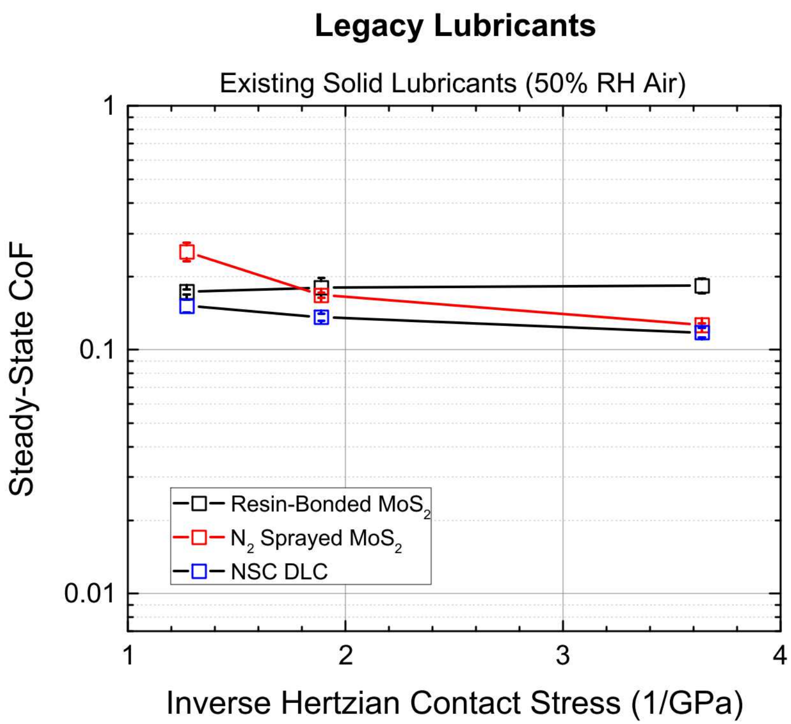
- Friction coefficient decreases as stress increases, typical of solid lubricant behavior
- Consider steady-state performance at each stress

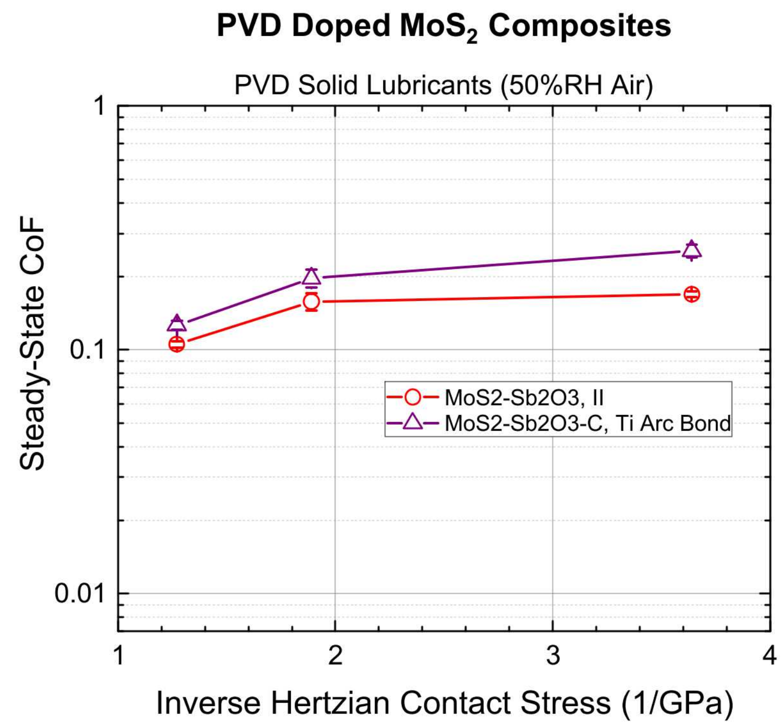
Friction vs Contact Stress in Dry N_2



- Impingement pure MoS_2 exhibits steady-state friction ≤ 0.05 at all stresses
- All doped MoS_2 composites exhibit lower friction coefficient than resin-bonded legacy coatings
  - several also exhibit lower friction than N_2 sprayed MoS_2 and DLC

Friction vs Contact Stress in 50% RH Air



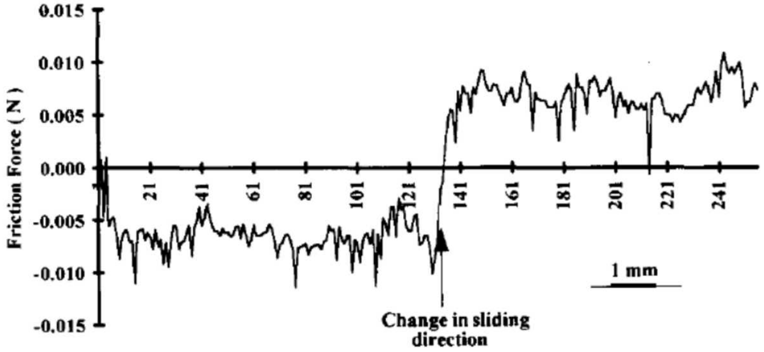


- All films exhibit increased friction compared to that in inert gas
  - the resin-bonded legacy film is the least impacted
- Several PVD coatings (MoS_2+Sb_2O_3+C) exhibit friction coefficient comparable to that of the legacy coatings

Superlubricity in Pure and Composite MoS_2

- Friction coefficient of pure (no C, O contaminants) MoS_2 synthesized and tested in UHV without exposure to air, ~0.002-0.005

C. Donnet, Th. Le Mogne and J.M. Martin, *Surf. Coat. Technol.* **62** (1993) 406

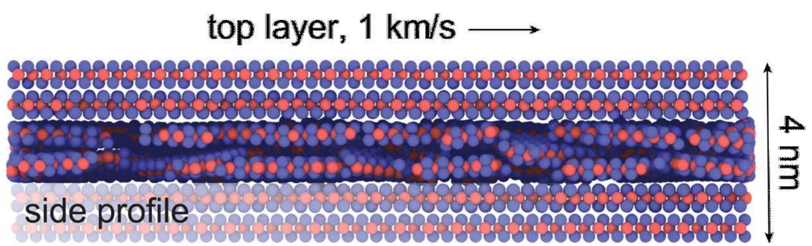


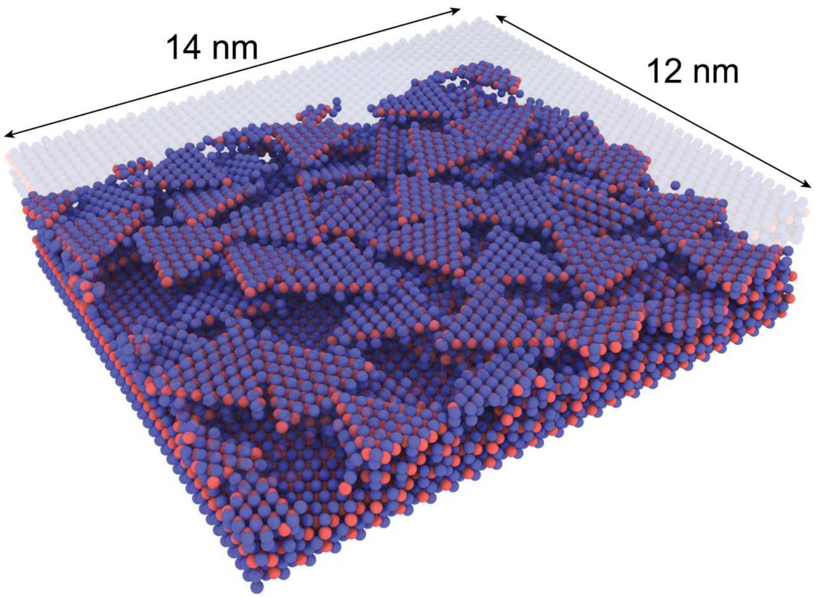
C. Donnet, et al., *Tribology International* **629** (1996) 123

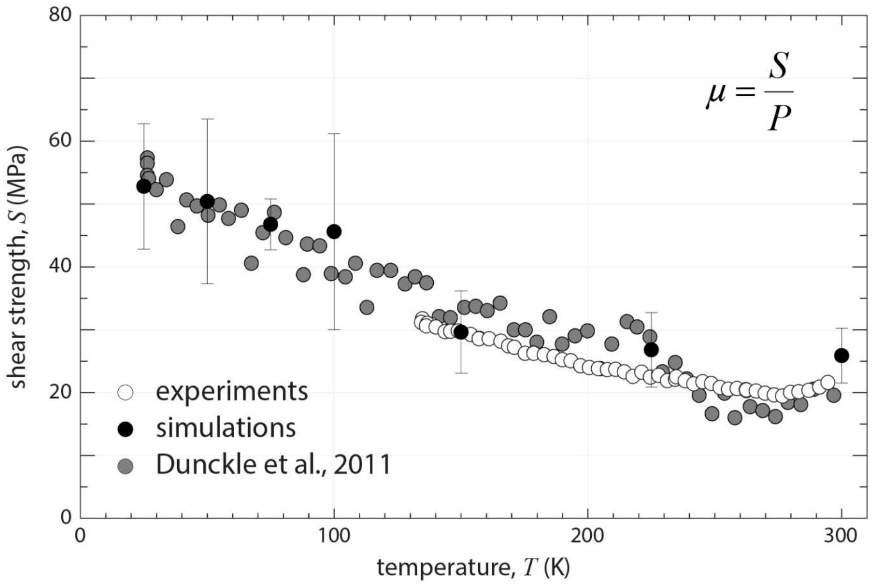
Atmosphere during friction	Average friction coefficient at cycle $N = 100$	Calculated shear strength S (MPa)
UHV	0.002	0.7
HV	0.013	4.9
d-N_2	0.003	1.1
at-Air	0.150	56.0

- In the present work, RF sputtered MoS_2-Sb_2O_3-C exhibited friction coefficients below 0.01 at similar stress, after shipping and testing in N_2
  - friction coefficient remains ~0.1 in atmospheres containing water vapor; not truly “environmentally agnostic”
  - characterization of aging behavior is ongoing

Physics-Based Friction Model for MoS_2





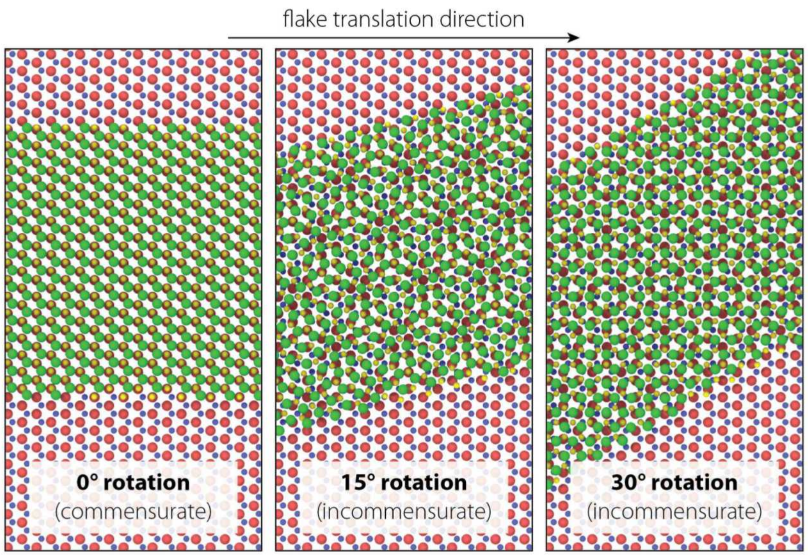


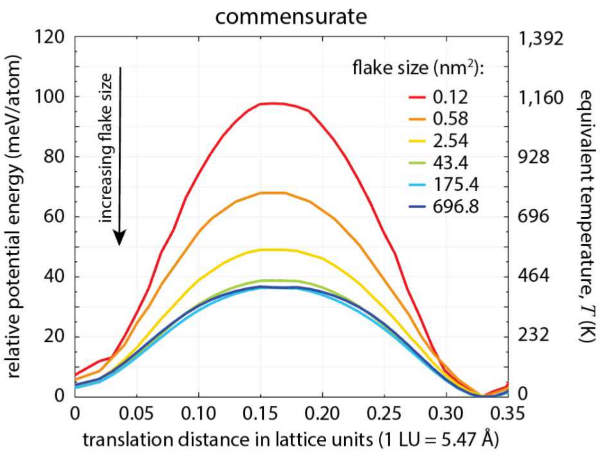
- Excellent agreement between experimental & simulation data for shear strength vs temperature
- Studying motion of individual flakes may provide insight into fundamental mechanisms

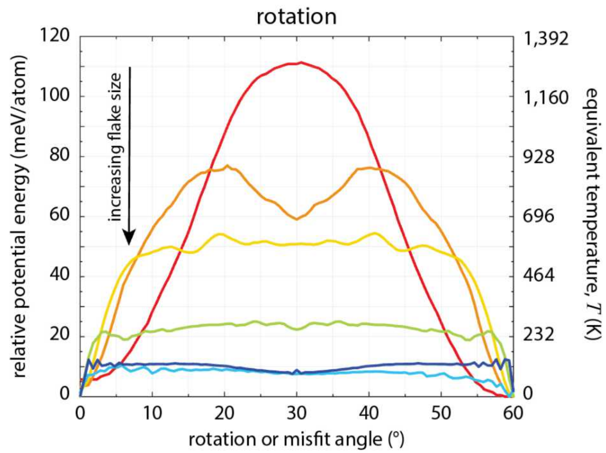
A basic understanding of the origins of friction in lamellar solids may permit compositing/doping to be used to mitigate environment and aging effects

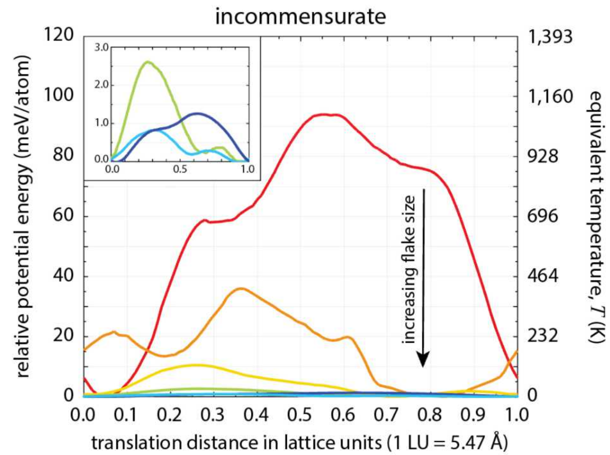
NEB Calculation of Energy Barriers

- Flakes of increasing size forced to translate/rotate and calculated required energies
- Energy barriers converge at larger flake sizes
- Commensurate sliding most energetically expensive route; incommensurate sliding **28X** less expensive

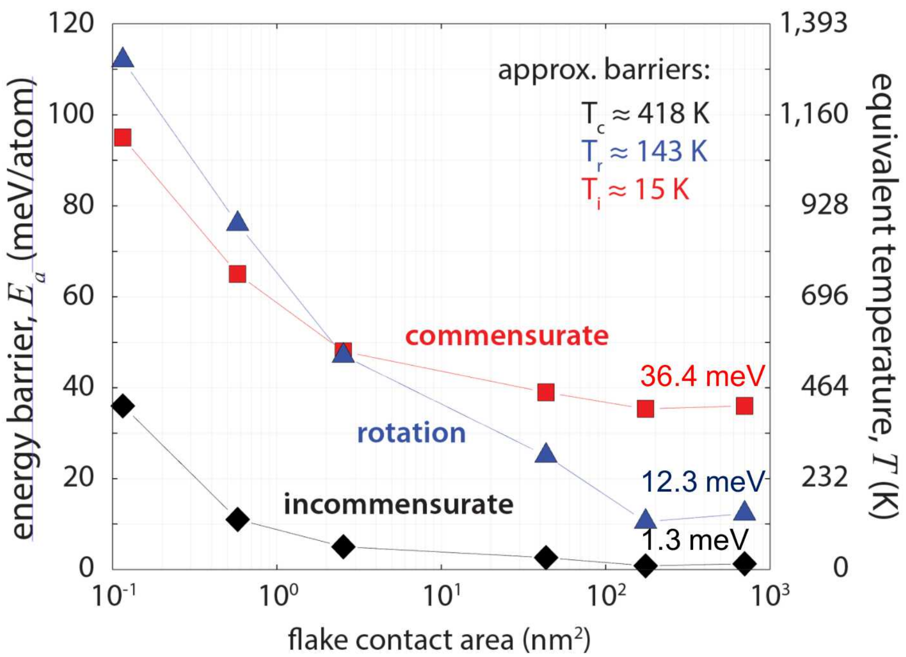








Converged Barriers & Analytical Model



- Model based on probability to overcome energy barriers to translation & rotation (Arrhenius)
- Expressed as inverse (1-exp) due to failure to thermally diffuse and slide under shear

The probability (p_n) and failure (f_n) to overcome a barrier:

$$p_n = A\exp\left(\frac{-\Delta E_n}{k_B T}\right)$$

$$f_n = 1 - p_n$$

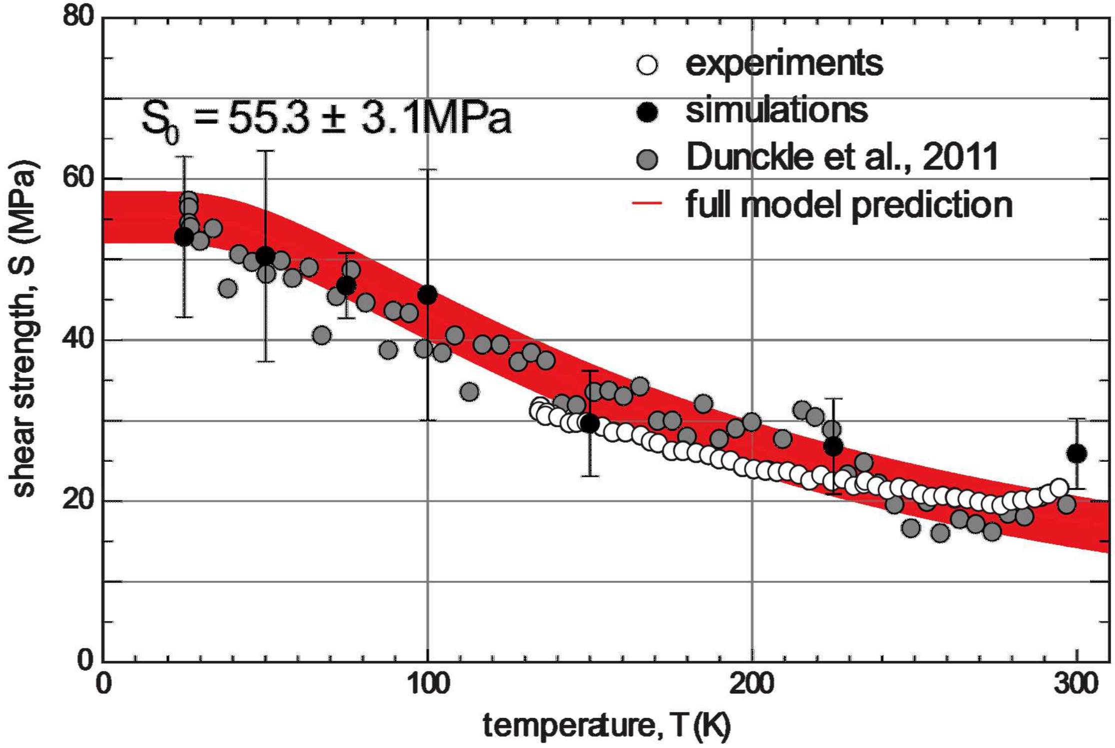
The probability to slide and fail to slide (friction):

$$p_{slide} = p_r p_i + f_r p_c$$

$$f_{slide} = 1 - p_{slide}$$

$$= 1 - \left(p_r p_i + f_r p_c\right)$$

Shear Strength via Barriers to Sliding



full model prediction:

$$S(T) = S_0\left(1 - \exp\left(-\frac{\Delta E_i + \Delta E_r}{k_B T}\right) - \exp\left(\frac{-\Delta E_c}{k_B T}\right) + \exp\left(-\frac{\Delta E_c + \Delta E_r}{k_B T}\right)\right)$$

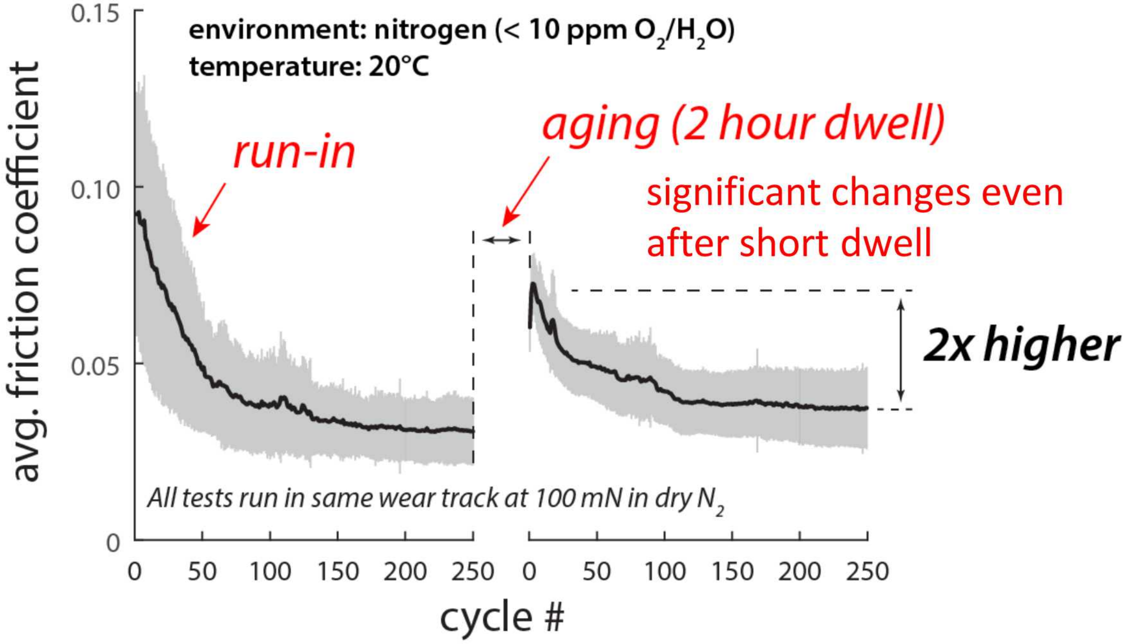
zero kelvin shear strength, S_0(T=0K)

successfully rotate; slide incommensurately

failure to rotate; slide commensurately

Concern after aging…"*re*-run-in"

Pure MoS_2 films ***after*** operating in inert atmospheres, ***still*** exhibit altered friction response after aging



A basic understanding of the origins of friction in lamellar solids may permit compositing/doping to be used to mitigate this and other aging effects

Conclusions

MoS_2–based solid lubricants exhibit environmental sensitivity and aging

- Requires robust mechanism designs to accommodate variability

Opportunities exist for improvements to solid lubricants for precision mechanisms in extreme environments

- PVD tools allow in-situ cleaning, thickness control, and tailoring of coating structure and chemistry on precision parts of complex shape
- It may be possible to synthesize new materials that minimize environmental heterogeneity, run-in, and susceptibility to aging

Phenomenological understanding of MoS_2 shear and alloying effects have already enabled design of superior materials

Physics-based shear models will enable optimization of solid lubricant structure/composition, and performance predictions

Acknowledgments

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