Plasma-Material Interaction and the Development of Liquid Plasma-Facing Components

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Outline of talk

- Introductory Remarks
- Challenges to Solid Plasma-Facing Materials (PFMs)
- A Brief Timeline of LMs in Nuclear Fusion
- Liquid Metal Surfaces I: Basics
- Liquid Metal Surfaces II: Applications
 - LM systems: integrating LMs in Fusion Devices
- Challenges with Liquid PFMs
- Summary and Outlook







Personal Background



- I've lived in many places since my childhood ۰
- Parents from Peru and Colombia, raised in Bogota, Colombia until I was ~ 8 years old
- Lived in Stavanger, Norway from 8-10 years old ٠
- Lived in Ciudad del Carmen, Campeche, Mexico, at 10 ٠
- Arrived at 11 to the United States and first lived in Shreveport, Louisiana finally settling in Los Angeles, California ۰



What inspired me in my scientific journey: self-organization in nature and Prigogine's dissipative structures?

ogi BS, in the chanical engineering, Physics minor sy California State Polytechnic University 1996 ystems

> Undergraduate Research in the Terms Experimental Tokamak (TEXT), 1994, 1995 Plasma turbulence and edge denaity fluctuations



Intel Corporation, Graduate Fellow Intern, Components Research Division 1996-1997

MS and Ph.D. in nuclear engineering, 2001 Ph.D. thesis. Plasma and radiation-surface ILLINUClear Fusioneand in the sard in the second of the s





Where I've been after my PhD...







Best of part of being a professor: watch my students succeed!













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Materials issues in Magnetic Fusion Energy

- Magnetic fusion energy presents many materials challenges, including:
 - High thermal heat fluxes
 - Sputtering/blistering of plasma facing components
 - Radiation damage
 - Low induced radioactivity
 - Chemical compatibility
 - Joining/Welding





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Stringent conditions in operational space of future plasma-burning fusion reactor environments

He in W is an issue for ITER, DEMO & FNSF.



14 MeV n's, high He/dpa up to 150 dpa for blankets up to 50 dpa for divertor

Can the liquid state provide an answer?

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PMI interactions are strongly coupled: plasma-surface and surfacesubsurface interactions (cont.)

Key question: How do these *PMI effects change for a liquid?*

Nature of the cascade and its damage plays a key role

* Wirth, Nordlund, Whyte, and Xu, Materials Research Society Bulletin 36 (2011) 216-222

A Brief Timeline of Liquid PFCs in Nuclear Fusion US Program

- The application of liquid metals in nuclear fusion reactors has a long history and many different efforts
- From fusion blankets to plasma-facing walls, liquids (metals, molten salts) have received the attention of many fusion scientists and engineers but most work remained basic/applied science

Liquid Metals I: Basics

- What is the difference between a liquid and a solid me
 - Very little difference in material density: solids are only 10
 - Key difference: time scale for atom mobility and macro sc
 - Another key difference: The surfaces between a liquid and

THE STRUCTURE OF LIQUID METALS

Progress in Metal Physics *B. R. T. Frost* Volume 5, 1954, Pages 96-142

1. INTRODUCTION

THE properties and behaviour of solid metals have been extensively investigated both from the point of view of the metallurgist and of the engineer. In recent years the substantial advances which have been made in the field of solid state physics have greatly improved the theoretical understanding of the subject. The study of metals in the solid state is relatively easy as the atoms are regularly arranged on a crystal lattice which can be examined in detail by means of x-rays and described in simple mathematical terms. A stage has now been reached at which many of the properties of solid metals can be quantitatively described in terms of the electron theory of metals or of dislocation theory.

In comparison with these studies of solids the investigation of the liquid state is still in its infancy. The experimental and mathematical problems are much greater because of the irregular atomic arrangement in the liquid state and the fact that in general the temperatures at which liquids exist are higher than those for solids. The study of gases is simpler than that of liquids since the interatomic forces in gases are much smaller and the ideal gas laws can be applied. From the practical point of view there is no lack of interest in liquid metals.⁽¹⁾ After the refining process, nearly all metals are melted and cast, either into a final shape or into a shape suitable for working in the solid state. Changes in the properties of the liquid metal may profoundly affect the soundness and quality of the casting. Increased interest is being shown in the use of liquid metals as heat exchange media in chemical and engineering plant and in nuclear reactors. The latter aspect has, in recent years, stimulated research on liquid metals and alloys in the United States and, to a lesser extent, the United Kingdom and Russia.

In liquid metals a phenomenon known as surface stratification occurs due to the charge state of the metal

- N.D. Lang and W. Kohn (1970's, Theory of Metal Surfaces), J. Penfold (1990's), many others...
- Peter Pershan (Harvard U.) and Stuart Rice (U. Chicago) were pioneers in developing modern techniques and models to study how liquid metal surfaces behaved
- The density of atoms near a surface would "stratify" or form a "layered" structure, this resulted in very interesting
 phenomena such as pre-melting, enhanced adsorption of impurities, and other effects that introduced implications to LM
 PFMs in fusion

Liquid Metals I: Basics

- How do liquid metals behave in a fusion environment?*
 - We want to know how fast does it erode (e.g. test stands with energetic particles and/or plasma)
 - Particles such as: D (fuel), He (from fusion reaction), impurities (metals, H, C, O)
 - We want to test its surface properties and how it interacts with substrates/structural materials
 - We want to also know how it behaves in complex environments such as a fusion device

PISCES-B

Li-DiMES

LIMITS

*both experimental test stands and computational modeling

Surface Properties: Evaporation vs Sputtering

- Evaporation is equally as important as sputtering yield as both inject wall materials into plasma
- Lowest melting point metals with attractive properties for fusion PFMs: Li, Sn and their alloys
- Sn has an evaporative flux many orders of magnitude lower than Li

Solid/Liquid/Gas thermodynamics: heat of fusion and heat of vaporization

Solid/Liquid kinematics:

surface binding energy – minimum energy to remove energetically an atom from surface

Indicent Particle Energy (eV)

Y. Waseda, S. Ueno, K.T. Jacob, J. Mat. Sci. Let, 8, (1989) 857-861.
 M.A. Abdou, A. Ying, N.B. Morley et al., Fus. Eng. Des. 2002

Erosion mechanisms in a liquid

- Erosion in a liquid is primarily dependent on the surface potential, U, of liquid atoms and the energy density deposition near the surface, F_D
- In the liquid state a large number of atoms are in motion in a given volume
- Enhanced erosion mechanisms are typically characterized by an increase in magnitude of the emitted flux as well as a decrease in the average ejection velocity of eroded particles¹⁻³
- Thermal sputtering and adatom ejection can increase with temperature

- 2. P. Sigmund and M. Szymonski, Appl. Phys. A 33 (1984) 141.
- 3. D.A. Thompson, Rad Effects, 56 (1981) 105.

D,He Liquid lithium -DIRECTION $Y = \frac{1^{\nu_{P}}}{2} \propto \frac{F_{D}}{2}$

The presence of oxygen on lithium surfaces

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Available online at www.sciencedirect.com

Paulos Engineering and Design 72 (2004) 111-119

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Fusion

Surface composition of liquid metals and alloys

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Sanaha National Laboratorics, Economere, CA 94337 -0900, USA

Available online 13 September 2004

Almiruci

In order to characterize the varfaces of liquids proposed for see as plasma facing materials in fusion reactors, the techniques of low-energy ion scalaring and data(incos) speciescopy have been used to coarrive the surface compacitizes of liquid Li. Ca. So, and a So-Li alloy in vacuum as a function of temperature. Oxygen is found to segregate to the variace of several metallic liquids. In the case of a So-Li alloy, Li also segregates to the liquid surface. Molecular hydrogen and its isotopes readily advorb on Li surfaces, bui not en trose of Un or Sn. Hydrogenic atoms al thermal energies can adsorb on both Li and Ca, bui no evidence has been found for hydrogen instepes residing at the surface of liquid Sa from the melting temperature to 800 °C. @ 2004 Elsevier B.V. All rights reserved.

Research: Liquid metals, Lithium, Gallicos, Tas, Hacingers, Desertant, Advertains, Sanface segregation, Low-energy ice acatering, Direct nexal spectroscopy. Parena surface interactions. Magnetic fasion reactors

I. Introduction

Liquid plasma-facing surfaces are being considered as an advanced materials option for magnetic fusion energy seactors, particularly in the divertor region where solid materials may not survive over long operating periods. Because liquid surfaces can be replonished, they offer the possibility of handling high power loads, recovering from plasma disruptions, and tolorating intense bombardment by neutrons and other plasma particles [1,2]. Three low-melting temperature metals, lithium, pallium, and tin (7,, = 180.5, 29.8, and 231.9 °C), have been selected as candidate liquid ma-

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terials, based on their plasma compatibility, reactivity with fasion neutrons, and thermodynamic properties Additionally, alkys made from these elements are also being considered, such as a binary alloy of Sn and Li.

The surface composition of a material can be different from its bulk composition, so a question of fundamental interest is: what is the composition of a plasma-facing liquid surface? This question is of considerable practical importance for the development of fusion technology since plasma-surface interactions are necessarily influenced by surface composition. In a first-step attempt to answer this question, the surface compositions of several candidate liquid materials have been examined using the low-energy ion scattering (LEIS) and direct recoil spectroscopy (DRS) methods. These techniques provide top-layer

Fig. 3. Variation in oxygen signal intensity on a lithium surface as a function of temperature. The intensity of ion scattering from O and most omission from Li wore monitored while the sample was hombunked with a 500 eV life. beam. Oxygen atoms appear more shundant on the liquid surface.

energy was periodically integrated for a short period and recorded along with the temperature of the sample. The results indicate that the surface oxygen level was higher when the sample was in the liquid phase. An obvious drop in surface oxygen coverage occurred near the freezing point of Li. In this case, the Li scattoring and receil signals remained relatively constant in both phases. As discussed below, these results point to thermodynamic segregation of oxygen to the surface of liquid Li.

Fig. 4. See energy spectrum of an air-expandi solid/Ga surface bombarded by 1 SeV He⁺ at 25 °C. Recoil emission of Ga⁺ and O⁺ is observed at the lower ion energies and He* scattered Sorn surface Ca and O atoms is observed at the higher energies.

ARIES Phase ~ 10⁻¹⁰ Torr. PH20 ~ 10⁻¹¹

3.2. Liquid Ga

3.3. Liquid Sn

800 °C.

An ion energy spectrum taken with a He* probe

beam of solid Ga soon after installation in the analysis

chamber is shown in Fig. 4. Distinct scattering and recoil peaks from both O and Ga are evident. The peak intensities, when scaled by the appropriate collision

cross-sections, indicate that slightly less than half of the

exposed sarface atoms were oxygen. Sputter treatment

removed the residual oxygen and produced a clean Ga

surface. Upon melting the clean sample, the surface be-

came mirror-like and an increase in the Oa LEIS signal

was observed, but no impurities were detected on the

surface at temperatures from 30 up to 500 °C. An ex-

ample of an ion scattering spectrum of the liquid Ga

The surface of unmelted Sn appeared to have a low

initial level of imparities when first installed in the anal-

ysis chamber. Small signals from H, O and possibly

S atoms present on the solid surface were present in

the ion energy spectrum, as shown in Fig. 6. When

melted, the spatter-cleaned Sn surface remained rela-

tively clean. As is illustrated in Fig. 7, no other ele-

ments, including hydrogen, were observed on the liq-

uid Sn surface from the melting tomperature up to

surface at 200 °C is shown in Fig. 5.

Fusion Engineering and Design 49-50 (2000) 127-134

www.elsevier.com/locate/luxenple

ALPS-advanced limiter-divertor plasma-facing systems

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Abstract

The advanced limiter-divertor plasma-facing systems (ALPS) program was initiated in order to evaluate the potential for improved performance and lifetime for plasma-facing systems. The main goal of the program is to demonstrate the advantages of advanced limiter/divertor systems over conventional systems in terms of power density capability, component lifetime, and power conversion efficiency, while providing for safe operation and minimizing impurity concerns for the plasma. Most of the work to date has been applied to free surface liquids. A multi-disciplinary team from several institutions has been organized to address the key issues associated with these systems. The main performance goals for advanced limiters and divertors are a peak heat flux of >50 MW/m². dimination of a lifetime limit for erosion, and the ability to extract useful heat at high power conversion efficiency (~ 40%). The evaluation of various options is being conducted through a combination of laboratory experiments,

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0920-3796-00;5 - say front matter © 2000 Elsevier Science B.V. All rights reserved. PE: \$0920-3796(88)00385-9

Detailed UHV surface analysis of lithium-based surfaces (solid and liquid) during ALPS program in U.S.

Review issue: Vol 72 in 2004 (Fus. Eng. Des)

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Temperature dependence of lithium sputtering

- Accounting for evaporation, ion-beam experiments identified temperature-dependent behavior of D and selfsputtering from liquid Li surfaces
- Experiments included: D saturation, oblique incidence, also He sputtering
- Many observed effects corroborated by linear plasma and tokamak device experiments

J.P. Allain, M.D. Coventry and D.N. Ruzic, Phys. Rev. B 76 (2007) 205434 J.P. Allain et al. Fusion Engineering and Design, 72 (2004) 93

Secondary Li ion sputtering fraction

- The secondary sputtered ion fraction was measured first from solid Li surfaces and then systematic studies with temperature
- Sn-Li experiments revealed the segregation of a pure Li layer on the surface, consistent with LEISS data by Bastasz et al.

J.P. Allain, M.D. Coventry and D.N. Ruzic, Phys. Rev. B 13 (2007) 056117R. Bastasz and W. Eckstein, 290-293 (2001) 19-24J.P. Allain et al. NIMB 239 (2005) 347; paper included MD simulations and stratification model for liquid Li

Fuel retention (e.g. D retention) in liquid Lithium

 D implanted at the lithium surface will lead to preferential sputtering of D atoms over Li

leading to Li sputter yield reductions of ~ $40\%^{1}$

- TDS measurements (Sugai, Baldwin, Evtikhin², Mirnov³ and others) show indirect evidence that D is implanted at the surface in solution with Li atoms based on their emission at temperatures (~ 400-500 C) lower than formation temp. for Li-D (T ~ 700 C)
- Both solid and liquid Li surfaces can retain 1:1 D:Li; *solid surfaces however must be replenished*

¹ J.P. Allain and D.N. Ruzic, Nucl. Fusion 42 (2002) 202.
 ² V.A. Evtikhin, et al. Plasma Phys. and Controlled Fusion, 44 (2002) 955.
 ³ S. Mirnov, et al. J. Nucl. Mater. 290-291 (2009) 87.

Coupling surface response codes with edge plasma codes and in-situ experiments

- In-situ PMI diagnostics (e.g. DiMES probe in DIII-D) already demonstrated the advantage of coupling:
 - In-situ PMI probe data
 - Computational modeling codes (edge, surface)
 - Off-line single-effect experimental data

J.P. Allain, J.N. Brooks and D.G. Whyte, Nucl. Fusion, 44 (2004) 655 D.G. Whyte, J.P. Allain et al. Fusion Eng Des. 72 (2004) 133-147

Stabilizing free-surface liquid Li layers

M.A. Jaworski et al. Nucl. Fusion 53 (2013) 083032

Li-DiMES experiments in DIII-D by Whyte et al. pointed to the importance of stability of liquid surfaces under tokamak conditions

D.G. Whyte et al. Fusion Engr Design 72 (2004) 133

- Li-DiMES experiments
- As indicated by Jaworski et al. "This usage of a porous substrate for stabilization of a free-surface liquid metal has been demonstrated in a diverted tokamak"

Persistence of Li layers at high temperatures indicated mechanisms linked with lithium/substrate interactions

- Significant reduction in Li erosion yield with temperature contributed to super-saturated region of implanted D (β = D/Li concentration ratio)
- Jaworski demonstrated stabilization regime for liquid Li layers on nanostructured or porous substrates
- Recent work by Allain et al. showing that Li layers persist at temperatures above 900 C in addition to the formation of fuzz nanostructure for He irradiation

T. Abrams, et al. Nucl. Fusion, 56 (2016) 016022

Allain and Ruzic, Nucl. Fusion 2002

M.A. Jaworski et al. Nucl. Fusion 53 (2013) 083032

What have we learned about liquid-lithium surfaces exposed to energetic D, He and Li bombardment?

- No significant difference in sputtering from the solid to liquid state of lithium when temperature is near melting point
- Non-linear increase in sputtering from liquid-Li when temperature is about 50% higher than melting point (accounting for evaporation)
- Two-thirds of lithium sputtered particles are in the charged state
- Implanted hydrogen leads to a ~ 40% decrease in *lithium* sputtering
- So far: liquid Li, Sn-Li, Ga and Sn show signs of erosion enhancement (particularly lithium) *with* rise in temperature
- Li-DiMES data shows near-surface ionization of emitted Li particles within ~ 1cm¹
- High retention of deuterium in liquid lithium (PISCES-B results by M. Baldwin et al.)²
- Critical to have 'stable' *flowing liquid lithium systems* due to: macro, micro and nano-scale oxide coverage; heat removal; etc...

¹ J.P. Allain J.N. Brooks, and D.G. Whyte, Nucl Fusion, 44 (2004) 655.

² M. Baldwin, R.P. Doerner, R. Causey, et al. J. Nucl. Mater. 306 (2002) 15

Model Integration of Various Beam-Target Interaction Physics in Computational Modeling Tools: HEIGHTS

A. Hassenein et al. Fus. Eng. Des. 2002

Vapor shielding mechanisms

- During the early stages of an intense power deposition on a target material (i.e. divertor, limiter), a vapor cloud from target debris is formed above the bombarded surface.
- This shielding vapor layer could be either beneficial or detrimental depending on application.
- Macroscopic particles (MP) emitted into the vapor cloud will significantly alter the hydrodynamic evolution of the vapor plasma.

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Liquid Metals II: Applications

- What are the different ways we may apply a liquid-metal as a plasma-facing material?
 - Inject a fast flow of liquid metal into the boundaries facing the fusion plasma
 - Deposit a low-melting point thin film, then melt it facing the plasma
 - Introduce the liquid metal slowly with some innovative techniques

Different mechanisms for establishing liquid walls*

• Gravity-Momentum Driven (GMD)

- Liquid adherence to back wall by centrifugal force.
- Applicable to liquid metals or molten salts.

- Electromagnetically Restrained LM Wall
- Externally driven current $(ec{J})$ through the liquid stream.
- Liquid adheres to the wall by EM force $\vec{F} = \vec{J} \times \vec{B}$

Fluid Out

- Magnetic Propulsion Liquid Metal Wall
- Adheres to the wall by $\vec{F} = \vec{J} \times \vec{B}$
- Utilizes 1/R variation in $\vec{F} = \vec{J} \times \vec{B}$ to drive the liquid metal from inboard to the outboard.

*M. Abdou et al. APEX Program 2000

Inlet velocity = 15 m/s; Initial outboard and inboard thickness = 50 cm

Toroidal width = 61 cm Corresponding to 10° sector
 Area expansion included in the analysis

The thick liquid layer:

- is injected at the top of the reactor chamber with an angle tangential to the structural wall
- adheres to structural wall by means of centrifugal and inertial forces
- is collected and drained at the bottom of the reactor (under design)

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Fast flowing liquid metal plasma-facing components

- Fast flowing liquid metal plasma facing components may prove to be an attractive alternative to handle both high steady state and transient plasma heat flux in a fusion reactor power plant, which would revolutionize control of the plasma-material interface.
 - Liquid metals continually replenish material and are self-healing, obviating lifetime concerns of solid materials, which erode with constant plasma bombardment.
 - In addition, certain liquids, e.g. lithium, can strongly improve plasma performance, leading to smaller, more economical reactor designs.
 - There are however, several important knowledge gaps in these systems, including:
 - managing the tritium fuel retention,
 - maintaining clean surfaces for reliable flow,
 - counteracting mass ejection forces,
 - determining operating temperature windows,
 - demonstrating helium ash exhaust

Application of liquid Li in divertor region of NSTX at Princeton Plasma Physics Laboratory

- LLD in NSTX at PPPL demonstrated two important results in the context of plasma-liquid interactions:
 - Mo influx disappears once lithium melting pt is reached during MHD events
 - No macroscopic amounts of lithium were injected from the LLD campaign
- Computational modeling tools linked surface response theory of liquid Li to experimental data in a tokamak
 - The importance of surface impurities such as oxygen and hydrogen (from water) became evident

Delivering liquid metal and providing a stable interface between the PFM and the edge plasma is part of emerging work in the field

Ghoniem and Williams, 2017

- Many concepts currently in development from macro-scale metal foam platforms for slow-flowing liquid metal to textured surfaces, e.g. see:
 I.E. Lyublinski et al., Nuclear Fusion, 57, (2017)
- Some groups are already examining adaptive PFC development testing hierarchical matrix systems that guide liquid metal in solid-based system at mesoscale; however much still at its infancy

New directions in porous metal self-healing structures for extreme environments such as magnetic fusion

- Adopting self-healing properties or *adaptive* properties for materials exposed to harsh environments
- Foam and porous materials are becoming an exciting direction for self-healing metal-based materials in extreme environments of heat, pressure and radiation
- Design of smart porous nuclear materials used in nuclear fusion reactors

Allain and Kapat 2016

Key Issue: Wettability of liquid metal to substrate, in this case liquid Li wetting of porous W substrate materials ("hybrid" liquid-solid interfaces)

- a) Left picture shows the complete lithium percolation of Li droplets immediately on porous W substrate
- b) Right set of pictures show the progression of lithium percolation after Li droplets is placed on porous W at 350°C, within ~10s.
 Wetting angles as Li percolates labeled.

E. Lang, A.Kapat, J.P. Allain, *Deciphering surface behavior and deuterium retention in tin-lithium-coated fuzzy tungsten substrates, NME* 12 (2017) 1352

Other concepts to deliver a lithium interface

**Understanding the trade-off between Fast-flowing liquids and static or slowflowing systems are the subject of current investigation

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Summary I: Advantages of LMs in fusion devices as PFMs

- Very high steady, and transient heat exhaust: 50 MW/m² exhausted from electron beam heating; also pulsed 60 MJ/m² in 1 μsec [22]
- Tolerable erosion from a PFC perspective: self-healing surfaces
- No dust generation
- Eroded chamber material from the main chamber transported to the divertor could be removed via liquid flow
- Neutron/dpa tolerance; underlying substrate would still have neutron-induced modifications, however
- Substrates below LM are protected from plasma-material interactions
- Liquid lithium specifically offers access to low recycling, high confinement regimes in certain surface temperature ranges
- Other liquid-metals such as Sn-Li alloys, Sn and Ga-based alloys could prove promising but their properties and behavior it's still not well understood
- Molten salts such as Flibe as a PFM could also provide alternatives, again more research is needed

Summary II: Challenges to LMs as PFMs in fusion devices

- Reliably producing stable LM surfaces and flows
- Understanding and controlling the LM chemistry
- Understanding liquid metal surface stability and interaction with plasma under relevant fusion reactor conditions (e.g. high-duty cycle, high temperatures, safety constraints, etc...)
- Acceptable temperature windows for specific integrated scenarios: choice of substrate/coolant able to provide for LM surface temperature control
- Fuel retention, recycling in liquid metals
- Corrosion issues involving large quantities of LM interfacing with substrate/bulk components at high temperatures
- Wetting vs dry-out effects asymmetric over substrate materials
- Neutron damage of solid-based substrate materials
- Understanding application of LM to a divertor vs the first wall
- Managing large He exhaust and tritium inventories

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Questions?

Extra Slides

Li-DiMES and computational modeling at the sheath

Evolution and lifetime of a lithium droplet (MP) moving in Li vapor

He diffusion in liquid Li

- For temperatures around 300 C, He diffusion in Li is about 5 x 10⁻⁴ cm²/sec
- This could be lower due to He cavity formation in liquid Li
- Atomistic simulation is a helpful tool to guide understanding on how liquid Li may pump He particles

Multi-phase and hierarchical design of the plasma-material interface: multi-scale porous materials

- Properties are determined by:
 - -Bulk mechanical strength, resilience and heat transfer
 - -Surface response to plasma exposure

• Steam generation from a thin porous graphene sheet. Steam plume is generated by harvesting thermal energy from sunlight exposure

T. Fujita, Sci. Technol. Adv. Mater. 18 (2017) 732

