SAND2022-8367C

Spectroscopic Evaluation of Tribologically-induced Chemical Changes in Zr-based Bulk Metallic Glass

Hsu-Ming Lien[1], Michael Chandross[3], and Filippo Mangolini[1,2]

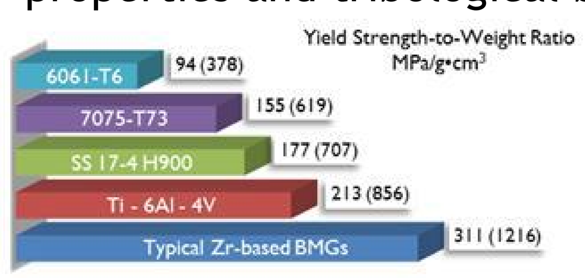
1. Texas Materials Institute, University of Texas at Austin, Austin, TX 78712 | 2. Walker Department of Mechanical Engineering, The University of Texas at Austin, TX 78712 | 3. Sandia National Laboratory, Albuquerque, NM 87123

The University of Texas at Austin Texas Materials Institute

Sandia National Laboratories

Introduction / Motivation

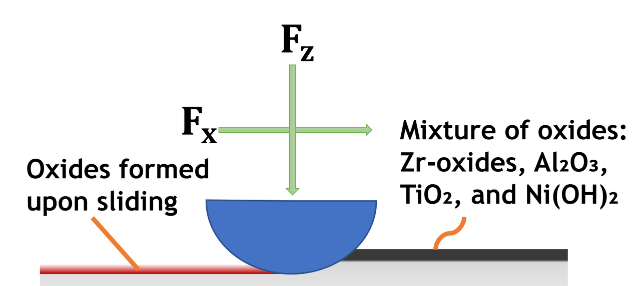
- Zr-based Bulk Metallic Glasses (Zr-BMGs): a class of promising structural materials owing to its high elastic limit (~2%)[1], yield strength-to-weight ratio[2], and high glass forming ability (GRA)[3].
- Origins of the promising tribological properties not yet understood because of the complex combination of (i) plasticity[4], (ii) structural relaxation and transformation[5], (iii) materials transfer, and (iv) atomic mixing at the surface[6]
- The determination of the surface chemistry and structures of BMGs and the evolution upon sliding is critical for correlating material properties and tribological behaviors.

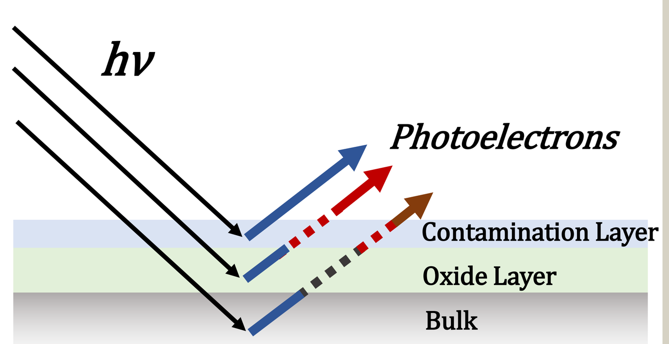


Aim of this work:
Evaluate the tribologically-induced surface chemical changes under different conditions

Experimental Methods

- **Material:** Vit 105 ($Zr_{52}Cu_{18}Ni_{15}Al_{10}Ti_5$)
- **Tribological Testing**
  1) Environments: ambient (60%RH) and low-humidity (~12%RH)
  2) Applied loads (N): 5 and 9.8
  3) Sliding speed (mm/s): 1, 3, and 10
  4) Number of cycles: 50
  5) WC ball (φ=2mm)
  6) Average Contact Pressure:
     - 1.34 GPa for 5N
     - 1.68 GPa for 9.8N





- **X-ray Photoemission Spectroscopy (XPS)**

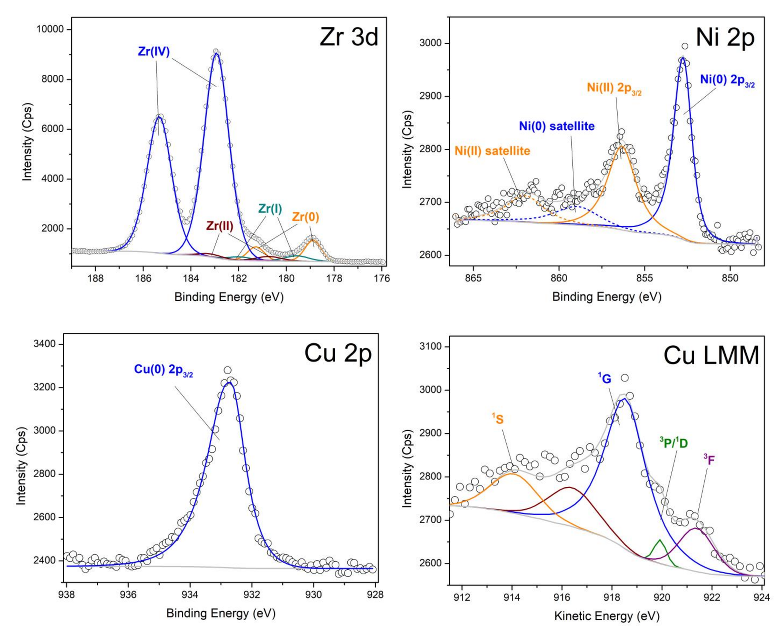
  The Three Layer Model (TLM) proposed by Asami *et al.* was applied to account for the attenuation caused by the adventitious contamination layer and the oxide layer on top of the bulk.[7]
- **Scanning Transmission Electron Microscopy (STEM)**
  1) Acceleration voltage: 200kV
  2) Energy Dispersive X-ray Spectroscopy (EDS)

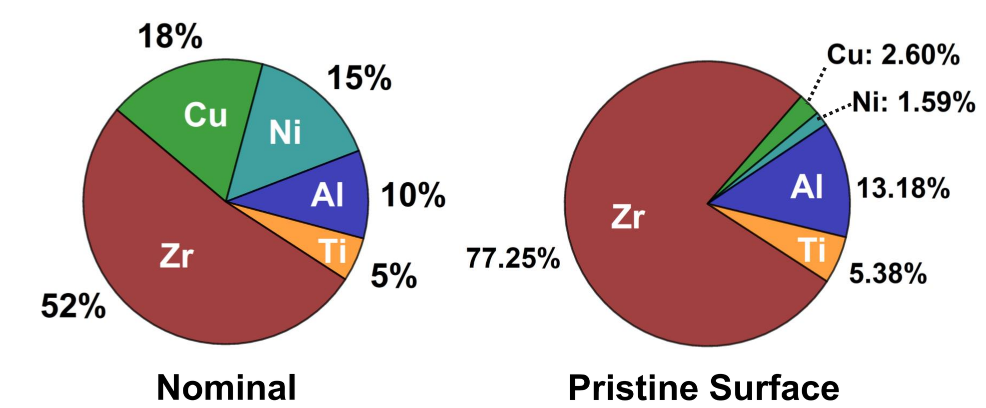
Acknowledgements

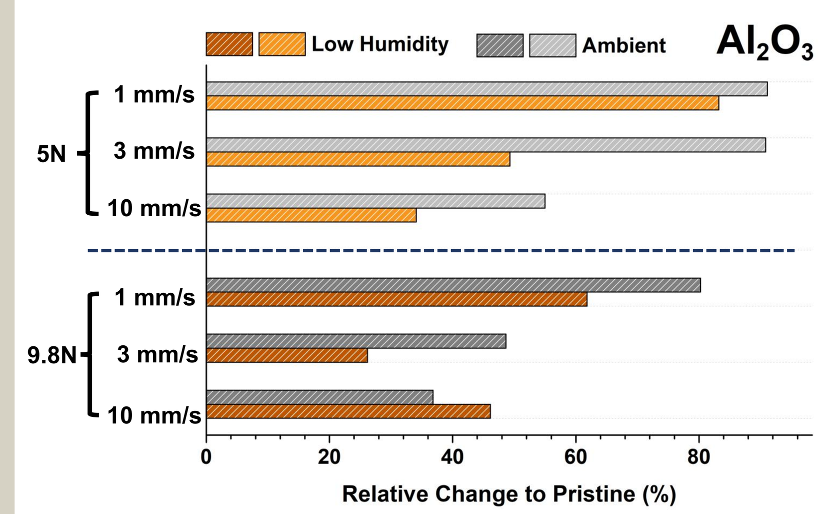
- The work in this poster is supported by Sandia LDRD grant, NSF CAREER Award, and the University of Texas at Austin.
- Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. SAND No. ____________

1. R Maaß and J.F. Löffler, *Adv. Funct. Mater.* **25** (2015), 2353-2368
2. M. Telford, *Mater. Today* **7** (2004), 36-43
3. A. Inoue and A. Takeuchi, *Acta Mater.* **59** (2011), 2243-2267
4. T.C. Hufnagel *et al.*, *Acta Mater.* **109** (2016), 375-393
5. J. Antonowicz *et al.*, *Phys. Rev.* B **93** (2016), 144115
6. M. R. Chellali *et al.*, *Phys. Rev. Lett.* **125** (2020), 205501
7. K. Asami and K. Hashimoto, *Corros. Sci.* **24** (1984), 83-97

X-ray Photoemission Spectroscopy (XPS)

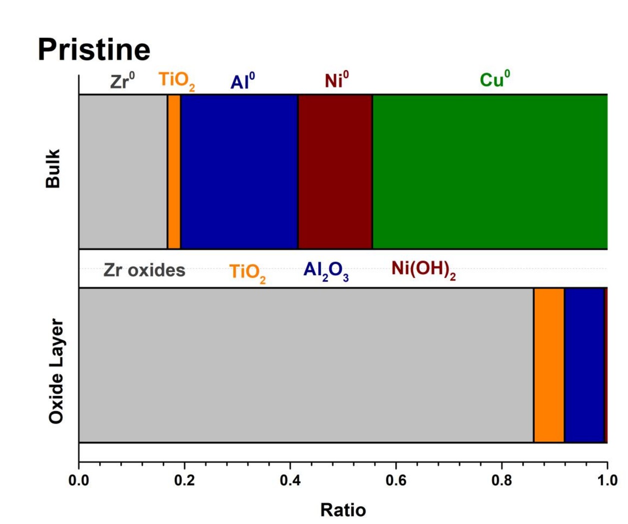


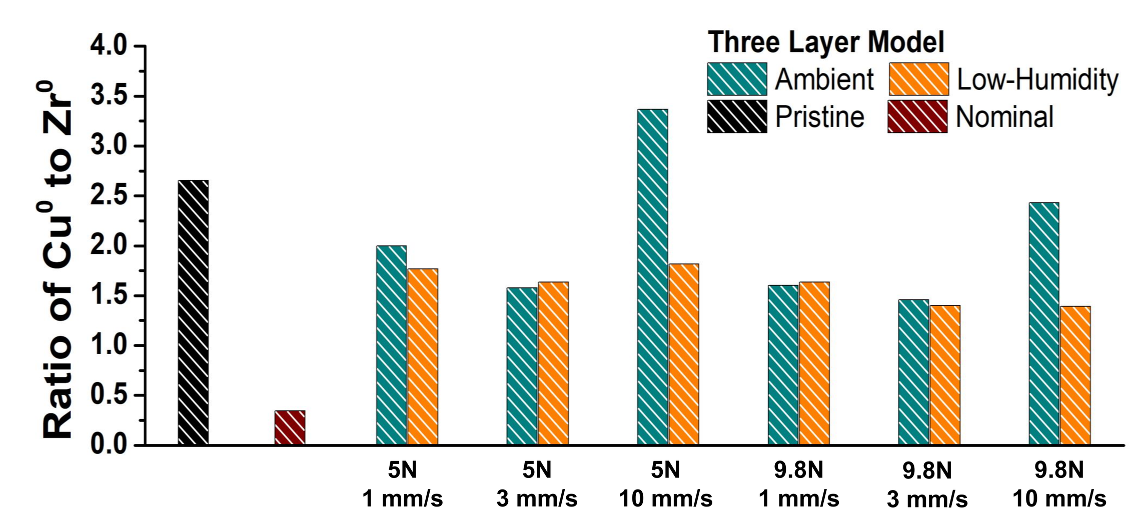


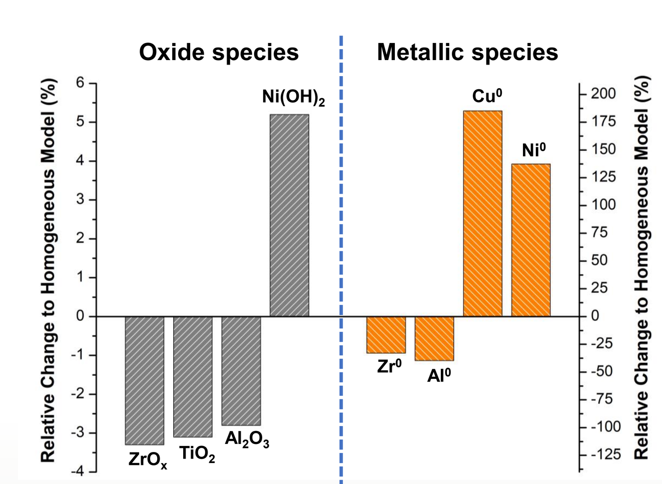


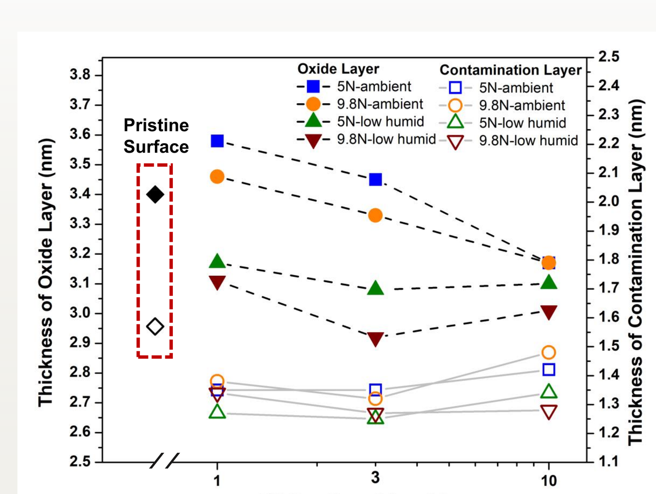
- Regardless of the applied load, sliding speed, and environment, the composition of the oxide layer changed upon sliding
- Much larger amount of alumina in the wear track
- Cu in metallic form and only presence in the bulk region
- The surface composition substantially deviates from the nominal one, especially for Cu and Ni
- No intermetallic phases observed upon sliding (peak positions and the corresponding chemical shifts suggesting the chemical states of oxide)

Three Layer Model (TLM)



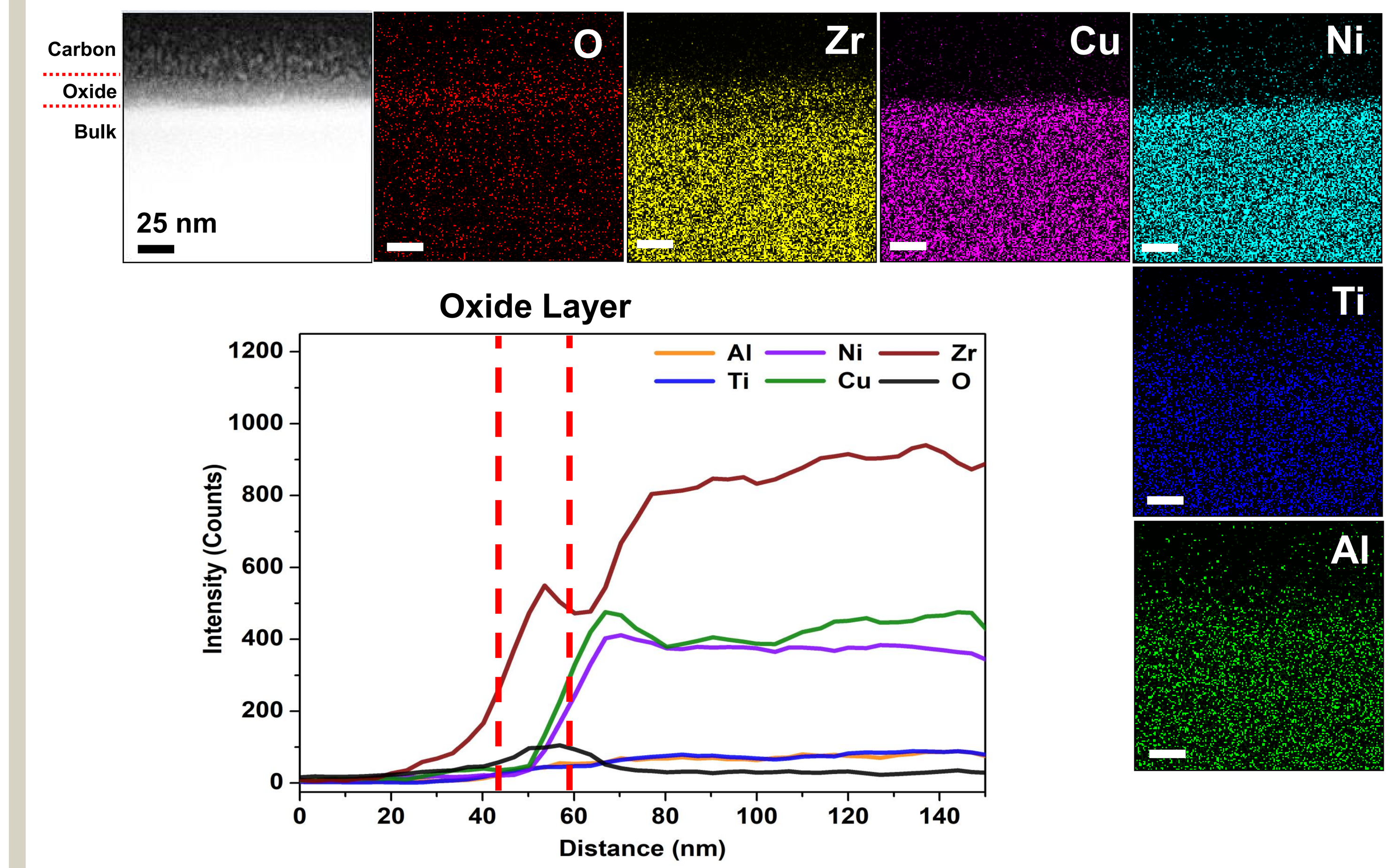




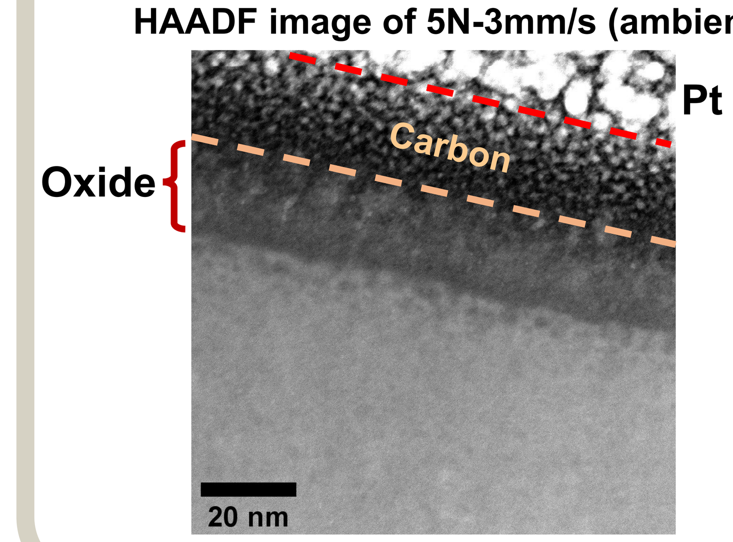


- **Composition of the layers:**
  1) Bulk composition deviating from the nominal one
  2) Substantial deviation of the bulk composition from the one computed assuming a chemically homogeneous volume probed by XPS
  3) Bulk region enriched with Cu^0 and Ni^0
  4) Oxide layer consisting of Zr-oxides, Al_2O_3, TiO_2, and minor Ni-hydroxide
  5) Positive deviation of Cu^0 to Zr^0 ratio from the nominal value suggesting the preferential oxidation of newly exposed metallic Zr in the bulk
- **Thickness of the layers:**
  1) Oxide layer thicker upon sliding in the aerobic conditions
  2) Adventitious contamination layer varies little with the environment

STEM EDS



HAADF image of 5N-3mm/s (ambient)



- Oxide layer primarily consisting of zirconium oxides
- Oxide-bulk interface rich in Cu and Ni but depleted in Zr.
- Homogeneous distribution of Ti and Al in both the oxide layer and the bulk
- No crystalline phases observed in the HAADF image

Conclusions

- The near-surface region of Vit105 has a composition that is significantly different from the nominal one.
- The TLM successfully predicted a Cu-rich region beneath the oxide layer, which is supported by the STEM EDS results.
- The native oxide layer is rich in Zr-oxides.
- No shear-induced crystallization observed via STEM
- Under low-humidity conditions, the bulk composition does not significantly vary with the tribological conditions.
- Under ambient conditions, the bulk composition depends on the strain rate; *preferential oxidation of Zr was more prominent at higher sliding speeds.*

Outlooks

- Molecular Dynamics (MD) simulations on the stress-assisted diffusion of Cu and Ni atoms in an amorphous glassy system
- Investigation of the atomic structures and the shear-induced structural transformations of Vit105 via synchrotron based X-ray Absorption Fine Structure (XAFS) techniques

  Accepted beamlines: (i) NSLS-II: 6-BM; (ii) APS: 13 ID-E

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.