

Novel Materials Discovery via High-Throughput Automated Tribological Testing of Thin Films

Tomas F. Babuska

Sandia National Laboratories

Kyle Dorman

Sandia National Laboratories

Justin M. Hall

Sandia National Laboratories

Manish Jain

Sandia National Laboratories

Michael T. Dugger

Sandia National Laboratories

Brandon A. Krick

Florida State University

Filippo Mangolini

The University of Texas at Austin

Frank W. DelRio

Sandia National Laboratories

David P. Adams

Sandia National Laboratories

Brad L. Boyce

Sandia National Laboratories

John F. Curry

`jcurry@sandia.gov`

Sandia National Laboratories

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Abstract

High-throughput automated testing offers accelerated ways to discover and characterize novel tribological materials. Here, we describe the design of a custom, fully automated, parallelized ball-on-flat reciprocating tribometer capable of performing upwards of 1000 friction experiments a day depending on contact conditions. Combinatorial physical vapor deposition was utilized to develop 448 Pt-Au alloy coatings spanning the full binary compositional range. Tribological performance was evaluated in both lab air and dry nitrogen environments, revealing multi-parametric dependencies of friction on composition, hardness, reduced modulus, surface roughness, and sputtered atom kinetic energy. In dry nitrogen, ultralow friction coefficients ($\mu_{ss} < 0.1$) were observed for Pt-rich coatings, with friction strongly influenced by both composition and the kinetic energy of Pt atoms during deposition. The low-friction behavior in dry N₂, attributed to the formation of tribofilms on Pt-Au, highlights the role of deposition conditions on surface tribochemical processes. Lab-air sliding experiments showed large variations in friction coefficients (~ 0.2 -1) across the entire compositional range and no trends with other modalities or modeled deposition atomistics. Benefits of adapting automation and/or parallelization to reduce operator and testing time were explored by calculating the total times for the presented data set. Systems with parallelized friction probes as well as automated systems are shown to reduce operator time by 99% and testing time by 90% compared to conventional serial testing. This work demonstrates the power of high-throughput tribological methods to generate large, multimodal data sets, paving the way towards self-driving laboratories in tribology that combine mechanistic insights and machine learning-driven materials optimization.

1. Introduction

The increasing prevalence and adoption of machine learning (ML) and artificial intelligence (AI) in materials science[1–4] has created a need for large, well-documented multi-modal data sets to train AI/ML algorithms. Data-rich fields such as tribology can leverage AI/ML techniques to predict functional properties of system-based material behaviors (*i.e.*, performance) like friction, wear and adhesion. For example, Lenz et. al. [5, 6] utilized Convolutional Neural Networks (CNNs) to determine the adhesion strength of thin films from both scratch adhesion[5] and Rockwell adhesion[6] experiments. Physics-informed multimodal autoencoders (PIMA) have also been used to identify differing permutations (*i.e.*, presence or thickness of adhesion layers or different substrates) of diamond-like carbon coating systems using only acoustic emission signals acquired during scratch adhesion experiments[7]. Though larger data sets are ideal, machine learning using small data sets with a gradient boosted regression tree (GBRT) methods found that an uncontrolled process variable, target conditioning time, strongly determines the initial coefficient of friction for molybdenum disulfide (MoS₂) coatings[8].

Providing large multi-faceted datasets to properly train and develop AI/ML models continues to be a challenge not only in tribology but many fields across materials science disciplines. Historically, experimental techniques have provided high-fidelity measurements on small sample sets (10s), yet exploration across large compositional spans has proven difficult because of limitations in synthesis

and testing methods or workflow bottlenecks. Increasing the throughput of material synthesis via processing methods such as high-throughput additive manufacturing[9–11], combinatorial physical vapor deposition,[12–15] and electrodeposition[16] has been demonstrated. High throughput coating creation via combinatorial PVD techniques has been used to create > 100 unique coating per deposition run for binary nanocrystalline alloy systems such as Cu-Ag[17] and Pt-Au[18]. The latter possesses attractive tribological properties including low friction and ultra-low wear rates[19]. Increased throughput of material property measurement techniques like tensile testing[20] have been achieved through a combination of automation and intelligent sample design[21] allowing for in-depth exploration of additive processes. For methods like nanoindentation, increased throughput is a combined result of automation and increased indentation speeds[22] allowing for mechanical mapping of multi-phase alloys using clustering methods on a data set containing one million indents[23].

In tribology, friction and wear reducing coatings are a promising field for application of high-throughput workflows[16] because accelerated experimental methods ranging from deposition (*i.e.*, process) to friction/wear (*i.e.*, performance) can be achieved. Functional properties like friction and wear are considered system properties as opposed to properties intrinsic to a material, as they depend on many factors such as environmental conditions [24], and loading conditions (*i.e.*, contact pressure, sliding velocity, sliding duration)[26], as well as the composition [25], and microstructural/mechanical attributes (*i.e.*, hardness/grain size and porosity)[27–30] of the materials in contact. Methods to accelerate friction and wear experiments, like increased speed, contact pressure, or temperature are difficult to employ, as they will inherently modify the tribological response of the interface. For example, Archard's wear law[31, 32] suggests that a wear test could be accelerated by increasing the contact pressure and proportionally reducing sliding cycles yet many materials systems have shown deviation from Archard behavior due to a change in the fundamental mechanism(s) driving wear[33–36]. An additional challenge is that for ultralow wear materials such as polytetrafluoroethylene-based (PTFE) nanocomposites[37–39], accurately resolving wear can require millions of sliding cycles [40]. Optimally designed high-throughput methods would overcome many of these challenges, enabling a robust characterization approach, revealing margins of material performance and potentially shedding light on results once seen as outlying from trends.

Beyond the challenges of developing infrastructure to acquire suitably large data sets for the purposes of training AI/ML models, there are also challenges in producing data at these scales that is findable, accessible, interoperable and reusable (FAIR). There is a notable deficiency in the amount of FAIR tribological data available[41]. Furthermore, when data is produced at larger scales, the quality of the data still remains the same, so methods for detecting and analyzing anomalous data across all modalities (*i.e.*, raw friction traces and surface topography data) becomes critical. These factors are all critical components in order to leverage integrated AI/ML models within automated high throughput systems to enable autonomous materials discovery, or “self-driving” tribological laboratories[42].

In this work, we discuss the development of a fully automated, parallelized high-throughput tribological test platform used to investigate combinatorially deposited Pt-Au thin films. Portions of this data set

have been used elsewhere to train AI/ML models capable of predicting the time dependent friction behavior of all Pt-Au samples tested in this study[43]. Pt-Au coatings are used as an exemplar system because of their unique mechanochemical behaviors that can be manipulated by changing ratios of Pt and Au alloying constituents. Previous work has shown the importance of Pt content[30, 44] on the resulting carbonaceous tribofilm formation driving low friction, yet to date, the experimentally tested composition range has been limited to only four compositions. Here, the full compositional span of the binary Pt-Au system is assessed to understand how different solute-solvent ratios, and their impact on microstructural and material properties, may enable differing mechanochemical responses.

2. Materials and Methods

2.1 Material Deposition via Combinatorial Physical Vapor Deposition

Combinatorial Pt-Au coatings were sputter deposited onto 4 sets of 112 (448 total) individual $10 \times 10 \times 3$ mm³, Ti-6Al-4V square coupons that were polished to an average roughness (R_a) < 20 nm. Samples were first cleaned ex-situ by sonicating coupons in a Brulin cleaning solution followed by a five-minute rinse in cascading ultrasonic deionized water and a five-minute cascading deionized water rinse. The rinsing process was repeated twice. Cleaned coupons were mounted on to a custom sample fixture which arranges the samples in a square array similar to [18]. A Kurt J. Lesker Inc. PVD-200 sputter-down system equipped with 7.5 cm TORUS™ Mag Keeper sputter guns, a turbomolecular pump, Ar mass flow controller, VAT Inc. throttling gate valve, and rotation stage was used to deposit all Pt-Au coatings. The base pressure of the process chamber was consistently below 5×10^{-7} Torr. Ultra-high purity Ar was introduced into the vacuum vessel and maintained at 10 mTorr for processing. Initially, a uniform, ~ 25 nm-thick Cr adhesion layer was deposited onto the Ti-6Al-4V substrates while rotating beneath a 99.95% pure Cr sputter target. Following the Cr adhesion layer, the samples were positioned for Pt and Au co-deposition. As described in ref [18], the rotation stage was set to a known, fixed position via encoder which placed the individual samples at different working distances from the two noble metal targets. The 99.995% pure Pt and Au targets were then pre-sputtered with the shutters closed for ~ 3 minutes in order to clean their surfaces of adventitious impurities. Afterwards, shutters were retracted and Pt and Au were co-deposited onto the stationary samples for 1300 seconds.

To span a near complete binary composition range in a few depositions (Fig. 1a), the angular position and power to each target were adjusted. The factory-set gun tilt position of 20° was used for some depositions, but targets were tilted away from this angle for experiments that sought reduced amounts of an associated metal species. The target – sample geometry is generally described in Supplemental Information. A detailed list of average powers, tilt angles, and discharge voltages used for deposition is included in Table A1 of the Appendix. Film thicknesses were measured on select samples which were masked at a corner using Kapton tape to produce a step edge. Utilizing a Sensofar S Neox 090 optical

profilometer, metrology demonstrated a range of thickness across different samples contained in a single deposition, typically spanning 0.3–1.1 μm .

The kinematic Monte Carlo-based program SIMTRA [45] was used to predict the binary metal composition, incoming kinetic energies of Pt and Au atoms, and the incident angle distributions. All compositions reported in this study derive from SIMTRA predictions but are expected to match true stoichiometry. The previous work [18] examined a nearly identical set of Pt-Au depositions and confirmed that SIMTRA-predicted compositions agree well with experimental values obtained by wavelength dispersive spectroscopy (WDS). The compositional range for each deposition is shown to have overlap with others (Fig. 1a) as well as the direction of the compositional gradient (*i.e.*, Pt decreases from bottom to top for all depositions) (Fig. 1b). Though not measured in this work, nearly identical deposition runs studied in [18] showed that other microstructural features such as density follow similar gradients to composition due to variations in local processing conditions (Fig. 2). The resulting 448 uniquely grown coatings allow for an in-depth exploration of the tribological properties across changing process conditions, microstructural variations, and binary composition.

2.2. High-Throughput Friction Testing and Property Measurements

Tribological testing was performed using a custom-made ball-on-flat bidirectional linear reciprocating tribometer with a robotically automated, parallelized and integrated design (RAPID) capable of simultaneous testing of six samples (Fig. 3). A detailed description of the tribometer components and operation can be found in the Supporting Information. Custom-made ball holders capable of holding three 3.175 mm diameter balls were developed to facilitate the automated testing of each sample in three separate locations. Automated ball holder loading, unloading and rotation between tests is performed using a 6-axis robotic arm (Mecademics, Meca500) with 5 μm point-to-point repeatability. The high-throughput tester uses a sample stage that holds 24 samples ($1 \times 1 \text{ cm}^2$) in sets of 4 per station and 3 platters of 24 or 48 ball holders placed near the robotic arm that can be swapped using a rotating assembly for a total of 144 available ball holders and 432 automated experiments before human intervention.

Development of the RAPID tribometer highlighted a critical flaw in approach – manual reloading of ball holders requires significant hands-on time and constrained the rate at which samples could be tested. For experiments with small sliding cycles or short strokes in the range of minutes, reloading ball holders begins to outweigh the duration of the experiments themselves. To overcome this limitation, a ball holder auto-changing system integrable with the RAPID tribometer was developed to enable the automatic unloading of used counterfaces and subsequent reloading of cleaned counterfaces. A detailed description of the auto-changing system can be found in the Supporting Information.

Tribological experiments consisted of three experiments on each sample with sapphire spherical counterfaces (Swiss Jewel) performed in both a dry nitrogen ($\text{O}_2 < 20 \text{ ppm}$, $\text{H}_2\text{O} < 20 \text{ ppm}$) glovebox and

lab air ($\sim 20\text{--}40$ RH%). A normal force of 100 mN, sliding speed of 2 mm/s and distance of 2 mm were used for each test. Longer 1000 cycle experiments were performed on each sample when tested in dry N_2 environments, whereas shorter 100 cycle experiments were performed in lab air. The differences for the cycle durations were due to the time required to reach steady-state sliding behavior which was longer in dry N_2 environments due to gradual development of tribofilms that lowered friction. The steady-state coefficient of friction is defined as the average of the last 100 sliding cycles across all three tests for dry N_2 environments and the average of the last 50 sliding cycles for lab air environments.

Hardness and reduced modulus were measured using a TI980 TriboIndenter (Bruker) with a Berkovich diamond tip. Samples were measured in batches of 56 using automated scripts and a custom sample stage. Nine displacement-controlled indents in a 3×3 grid with $10\text{ }\mu\text{m}$ spacing were performed on each sample to a depth of 40 nm under quasi-static conditions using a profile consisting of a 5 second loading period, 2 second hold at peak load, and 5 second unload. The hardness and reduced modulus were calculated using the Oliver and Pharr method[46] and the tip-area function was measured after each 56 samples on a fused-quartz reference sample. The tip-area function was fit using a 6th order polynomial and was valid down to 25 nm. It should be noted that the tip area function did not significantly change between sets of 56 samples.

Surface roughness (S_q) was characterized by atomic force microscopy (AFM, Bruker Dimension FastScan Pro) equipped with FastScan-C silicon nitride (SiN) cantilevers (nominal spring constant $k_c = 0.8\text{ N/m}$, tip radius $R_t = 5\text{ nm}$). Prior to use, the actual spring constant for each cantilever was determined via the thermal fluctuation method, yielding values between 42 N/m and 56 N/m. All measurements were performed in PeakForce Tapping mode with an applied force (F_a) of 10 pN to minimize tip-sample interaction. Topography scans were acquired over a $1\text{ }\mu\text{m} \times 1\text{ }\mu\text{m}$ area at a scan rate of 3 Hz and a resolution of 256×256 pixels. To maintain consistent imaging quality and minimize tip wear, each probe was replaced after 56 scans ($58 \times 8 = 448$ total scans). Scan parameters, including setpoint and drive amplitude, were optimized using the Bruker ScanAsyst tool to enable automated acquisition across the Pt-Au sample surfaces. Topography data were processed using NanoScope Analysis software; images were flattened to correct for sample tilt or stage bowing prior to S_q calculation.

3. Impact of Automation Method on Testing and Operator Time

Two primary methods were considered to increase the throughput of tribological experiments: (1) The sequential automation of a single measurement on the same sample (as with nanoindentation[22]) or on multiple (*i.e.*, tensile testing[20, 21]) and (2) the parallelization of the same measurement to allow for testing of multiple samples simultaneously. To highlight how automation and parallelization methods impact both operator time and tribological test time, we use estimates of generic test operations (*i.e.*, entry into glovebox, sample installation and program execution) based on dry N_2 and air data sets and show that automated parallel testing can decrease testing time by 90% and operator hands-on time by 99% (Fig. 4).

The test methods are described as the following: **Manual serial** testing is defined as a single experiment run on a single sample that requires an operator to interface with the equipment between individual experiments to either change the sample or equipment setup (*i.e.*, rotate ball holder, replace ball holder, replace sample stage and ball holder platter after 24 samples are tested). **Automated serial** testing is the same as the manual serial testing method but utilizes automation methods (*i.e.*, robotics, gantries, fixturing) that allows for at least two or more samples/setups (*i.e.*, ball holders) to be automatically changed between measurements without operator intervention. **Manual parallel** testing is the same as the manual serial testing procedure but replicated such that two or more measurements, either on the same sample or on separate samples, can be simultaneously performed. **Automated parallel** testing (Fig. 3) is defined as an experimental platform that can simultaneously perform two or more experimental measurements and allows for at least two or more samples/experimental setups to be changed without operator intervention.

The total time in days required to finish testing 448 samples (Fig. 4a) and total hours of operator time (Fig. 4b) were used to compare each method to demonstrate gains in efficiency of adopting parallelization and automation. A detailed description of the assumptions used for the time calculations in Fig. 4 are in the Supplemental Information. The results from the calculated times in Fig. 4 highlight the advantages and disadvantages of using automation and/or parallelization methods. It is unsurprising to find that the manual serial testing method (*i.e.*, traditional experimentation) requires the most operator time (13.2-18.96 hours) and test days (10.93–89.60 days) regardless of test length/environment. The strength of adding automation to an experimental process is in reducing operator time, with a ~ 97–98% reduction in operator time for an automated method over a manual process. For this test matrix, it could be completed with as little as 0.24 hours (or 15 minutes) of operator interaction by incorporating automation methods. Though adding automation decreases both the testing time and operator time, longer experiments benefit more from automation, supported by a 64% decrease in testing time between manual testing and automated serial for the dry N₂ experiments compared to a ~ 15% decrease for lab air. The addition of parallelization, regardless of automation, improves the testing time by 75–82% (depending on test length/environment) over manual serial testing. Optimization of both testing and operator time for both environments is achieved through the addition of automation to a parallelized system, with a 98–99% reduction in operator time compared to manual serial testing and an 83–90% reduction in testing time.

While the reductions in testing time and operator time have been shown to be reduced by 90–99%, optimizations can still be made that would accommodate a wider range of test conditions, sample sizes, and incorporation into self-driving workflows. For example, further automation efforts related to supplying the tester with sample plates and ball holders would enable testing multiple material sets with only one single operator setup. Additionally, for short duration experiments, the rate at which reloading can be accomplished becomes important to ensure that test time is not increased due to sample resupply. Sample resupply is also linked to considerations of sample quantities, where intermittent delivery of small sample quantities effectively delay automation and preclude capitalizing on overnight testing opportunities. Environmental control and purge times must also be considered for short tests as

inefficient transfer methods can prolong test durations due to slower environmental turnover. Other considerations not explored here are factors including high throughput ways to calibrate and/or validate test equipment, replace parts, swap to other modules (*i.e.*, varying load ranges) and perform maintenance without significant downtime.

Future directions include coupling high-throughput tribometers with other tribologically relevant techniques such as nanoindentation, surface profilometry, AFM etc., to create an automated tribology laboratory [42, 47–50]. These automated multi-modal experimental capabilities could then be coupled with multiple AI agents trained to interpret results in real time and dictate future experiments, rapidly accelerating our ability to explore fundamental mechanisms and discover novel materials.

4. Automated High-Throughput Exploration of Pt-Au Coatings

The friction behavior of 448 combinatorial PVD processed Pt-Au coatings was measured using the RAPID tribometer (Fig. 3) in lab air and dry N₂ to understand the effects of process variables and material properties on friction response. While lab air environments tend to have higher coefficients of friction than dry N₂, creating relationships between coating microstructure, process conditions and friction is difficult due to the large amounts of data. As the friction behavior in dry N₂ demonstrates, low friction occurs on Pt-Au alloys due to the mechanochemical formation of a lubricious tribofilm[24, 26], previously shown to be dependent on the coating hardness[30] and coating composition[44].

To better understand relationships between friction coefficient and material properties, steady-state coefficients of friction (μ_{ss}) are calculated from the cycle average friction traces and compared to composition (Au at. %), hardness (H), reduced modulus (E_r), and average surface roughness (S_a) for lab air (Fig. 6) and dry N₂ (Fig. 7) environments. In a lab air environment, steady state friction coefficient varies from $\mu_{ss} = 0.2$ – 1.0 across the entire compositional range (Fig. 6a), with a notable outlier below $\mu_{ss} = 0.2$ near Pt_{0.58}Au_{0.42}. While it appears that coatings with higher Au content exhibit more compositions with lower μ_{ss} , the large amount of variation in μ_{ss} with composition makes it difficult to draw links between composition and friction behavior in air. When comparing average friction coefficient to the coating hardness (Fig. 6b), results show no significant relationships though the lowest coefficients of friction are observed for softer coatings. Moore *et al.*, [52] suggested that friction coefficient for metals should linearly decrease with hardness, yet they assumed the formation of a thick surface oxide which is not present on Pt-Au. Following Amontons's law[53], the coefficient of friction under a constant contact pressure decreases if the material's shear strength decreases. In fact, softer metals such as Ag are used as lubricants[54] as they have lower shear strengths. For nanocrystalline metals like those studied here, low friction for Pt-Au coatings in air was originally attributed to grain-boundary sliding for alloy compositions that have grain boundary segregation of Au[19]. Below a critical stress, the grain size will not undergo coarsening[27, 28] and low friction can be maintained. In this study, we did not explore the variety of microstructures that can occur across the entire compositional range, and our results suggest that the chosen contact conditions were above any critical thresholds to promote low friction for most of

the tested coatings. Furthermore, upon comparing μ_{ss} to reduced modulus (Fig. 6c) or average surface roughness (Fig. 6d), no obvious dependence was observed. It should be noted that in this work other surface roughness parameters such as skewness, kurtosis, maximum height, mean height, waviness, or power spectral densities were not investigated yet it is possible other features of the surface topography scans could be related to trends in friction due to variations in roughness across length scales[55]. More detailed analyses of roughness by leveraging AI/ML methods with spectral analysis[56] may be able to elucidate connections between roughness and friction that are lost due to scalarization.

Although the friction behavior for Pt-Au coatings did not show any obvious trend with composition in lab air, results for dry N_2 (Fig. 7a) do demonstrate a relationship between dry N_2 μ_{ss} and Pt content. The lowest coefficients of friction, with some approaching $\mu_{ss} = 0.02$ or below, occur for the highest Pt composition coatings consistent with previous studies on Pt-Au alloys having up to 10 at. % Au[24, 26, 30, 44] due to the formation of lubricious carbon tribofilms. Carbon tribofilms in and along the wear tracks were observed for most of the coatings that were not predominantly Au rich (*i.e.*, deposition 4), though not characterized for this study. While the coefficient of friction can vary for every composition, the results show that low friction ($\mu_{ss} < 0.1$) can be achieved for coating compositions up to 80 at.% Au and 20 at.% Pt. For coatings with Au composition above 80 at.%, friction coefficient rapidly increases with increasing Au content and peaks for the highest Au containing films. For the coatings that experience carbon tribofilm growth, previous work by Edwards *et al.*[44] showed that the structure of the tribofilms were similar to other amorphous carbon coatings, changing with composition and highly influencing the coefficient of friction. To further understand why there is a spread in friction for similar compositions, μ_{ss} is compared to nanoindentation hardness (Fig. 7b), reduced modulus (Fig. 7c) and the average surface roughness (Fig. 7d). As with lab air environments, average surface roughness shows no clear correlation with μ_{ss} for all depositions. For depositions 1–3, μ_{ss} tends to decrease with increasing hardness with the lowest μ_{ss} values occurring for the hardest, Pt rich deposition (deposition 1). Notably, there is a deviation for the most Au-rich samples (deposition 4) where hardness and friction coefficient show no correlation. A similar relationship is observed between μ_{ss} and reduced modulus, with high moduli coatings having lower μ_{ss} and the Au rich deposition 4 showing no relation. The observed correlations between hardness/reduced modulus and μ_{ss} could be due to local changes in contact pressure with harder/higher modulus coatings exhibiting an increase in contact pressure. With increasing contact pressure and a constant shear strength, as described by Amontons's law and shown for other solid lubricants like MoS_2 [57], the coefficient of friction will decrease. It is also possible that the structure of the amorphous carbon surface film is changing due to changes in hardness and composition.

The underlying mechanisms driving changes in the average coefficient of friction, as shown in Figs. 6 and 7, are unclear even with the large amounts of data generated on 448 coatings. While coatings properties such as hardness, reduced modulus, and roughness are not clear indicators affecting average friction, ML models developed by Brown *et al.*[43] using nanoindentation values as the sole input were able to predict the time-dependent friction behavior of Pt-Au. Composition influences friction in dry N_2

environments, though the variation in friction is large for any given composition. Jain et. al., [58] showed that compositional modulation may be a strengthening mechanism (*i.e.*, increased hardness) for Pt-Au alloys within the miscibility gap due to spinodal decomposition which may account for variations in hardness at similar compositions. The films in this work are similar to those considered by Jain et. al., and could have the same underlying mechanisms driving variations in friction and hardness. As shown in Fig. 2, the average kinetic energy of Pt and Au atoms incident on a growing film have varied in addition to net composition. To understand the impact of process conditions on friction, μ_{ss} is plotted against composition with each point color coded by the calculated Pt and Au atom energy from SIMTRA (Fig. 8). In lab air environments (Fig. 8a), including Pt atom kinetic energy does not appear to explain any of the variations in friction. Interestingly, for dry N₂ environments (Fig. 8b) the addition of Pt atom energy shows that for any given composition, the friction coefficient will decrease with increasing Pt atom energy. The lowest coefficients of friction are shown for a range of compositions when deposited with high energy Pt (average approaching 3 eV in this study). Color coding friction as a function of composition by the average Au atom energy for both lab air (Fig. 8c) and dry N₂ (Fig. 8d) does not distinguish variations in friction, suggesting that the kinetics of incoming Pt atoms, and not Au atoms, are the driving factors behind Pt-Au friction. Desai et. al.[59] used a feedforward neural network (FNN)-based analysis of 13 different modalities for a similar Pt-Au system and found that higher kinetic atom energies generally resulted in high film densities and better mechanical/electrical properties. The results shown in Fig. 8b highlight the importance of tuning the process conditions for a given coating to not only target a desired compositional range but also considering the energy of deposition. In this work, we do not attempt to understand the underlying mechanisms for why energy may affect friction but instead focus on highlighting the power that high-throughput methods bring in exploring the process, structure, property, and performance space of a material.

5. Conclusions

In this work, we describe experimental methods for high-throughput, automated tribological testing of a large set of Pt-Au thin film samples fabricated via combinatorial PVD methods. An in-depth discussion on the role of automation vs. parallelization on the total test time and operator time highlights the importance of adding both automation and parallelization to tribological systems for maximum efficiency. By adopting high-throughput methods, a 99% reduction in hands on time and 90% reduction in testing time can be achieved over conventional manual serial testing methods. Using a robotically automated high-throughput tribometer, a large sample set of 448 Pt-Au coatings was studied to understand the effects of composition, hardness, reduced modulus, surface roughness, and kinetic energy on the resulting friction behavior in lab air and dry N₂ environments. Results show that in lab air environments, friction only weakly follows trends in composition with the lowest values occurring for the highest Au content coatings. In dry N₂, existing relationships between friction, composition, and hardness were expanded to the entire binary compositional span, demonstrating the importance of composition in driving low friction. The variation observed in friction for similar compositions can be

explained by the Pt atom kinetic energy used to grow each coating, with higher energies resulting in the lowest coefficients of friction for any given composition.

A crucial highlight of this work is the opportunities that high throughput testing enables when discovering novel materials or compositions that may have never been explored by conventional serial platforms. By testing 448 compositions, new performance criteria such as frictional variance of similar Pt-Au compositions can be determined and interesting outliers can be explored. Additionally, this work shows how fundamental mechanistic frameworks may change when data sets increase in size and fidelity. Previous work by these authors on four Pt-Au coatings showed that hardness is an important aspect governing mechanochemical phenomena, yet this results shown here suggest a nuance when using hardness in the same framework. While combinatorial samples are distinct from those produced with fixed targets, a separate consideration for this work is that friction is reduced from a time dependent behavior to a scalar value, losing valuable trends that could be analyzed with future AI/ML models. The promise of incorporating AI/ML using this data set has been shown in previous work by Brown *et.al.* [43] where the evolution of cycle average friction coefficients was predicted. Future work on increasing the throughput of experimental platforms, self-driving labs, data ontologies, FAIR data, and anomaly detection methods will enable larger, digestible tribological data sets for AI/ML methods. These future efforts will be critical in implementing future work on data-driven decision algorithms for autonomous decision making. Future work will focus on understanding the fundamental mechanisms leading to lower friction for coatings grown at higher kinetic energies, as well as a deeper understanding of carbon tribofilm formation and resulting changes in tribofilm structure as a function of alloy properties.

Declarations

Author Contribution

T.B. wrote the main manuscript and prepared figures. K.D. and D.A. performed depositions and did SIMTRA analysis. J.H. M.D. T.B J.C and B.K. designed the custom experimental platform. M.J. performed AFM experiments and analysis. F.M. and J.C provided conceptualization of high throughput tribology workflows. F.D performed nanoindentation, analysis and general data interpretation. B.B. and J.C provided funding and data interpretation. All authors edited and reviewed the manuscript.

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Data Availability

The DOE will provide public access to results of federally sponsored research in accordance with the DOE Public Access Plan.

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Figures

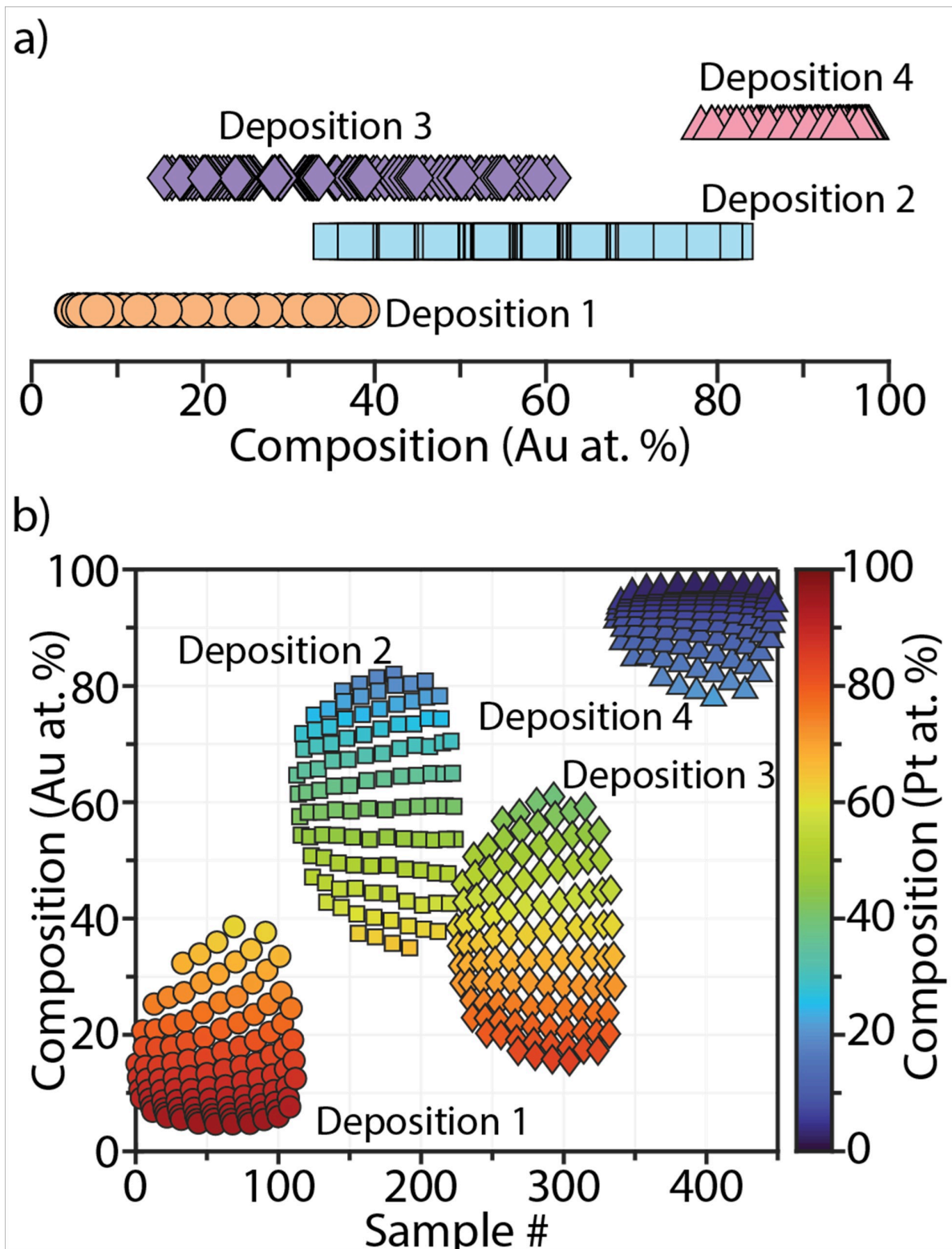


Figure 1

(a) The calculated composition range by SIMTRA of the four different sets of thin films expressed in Au at. % and (b) the composition gradient for all 448 samples across all 4 depositions (112 samples each) created by combinatorial deposition.

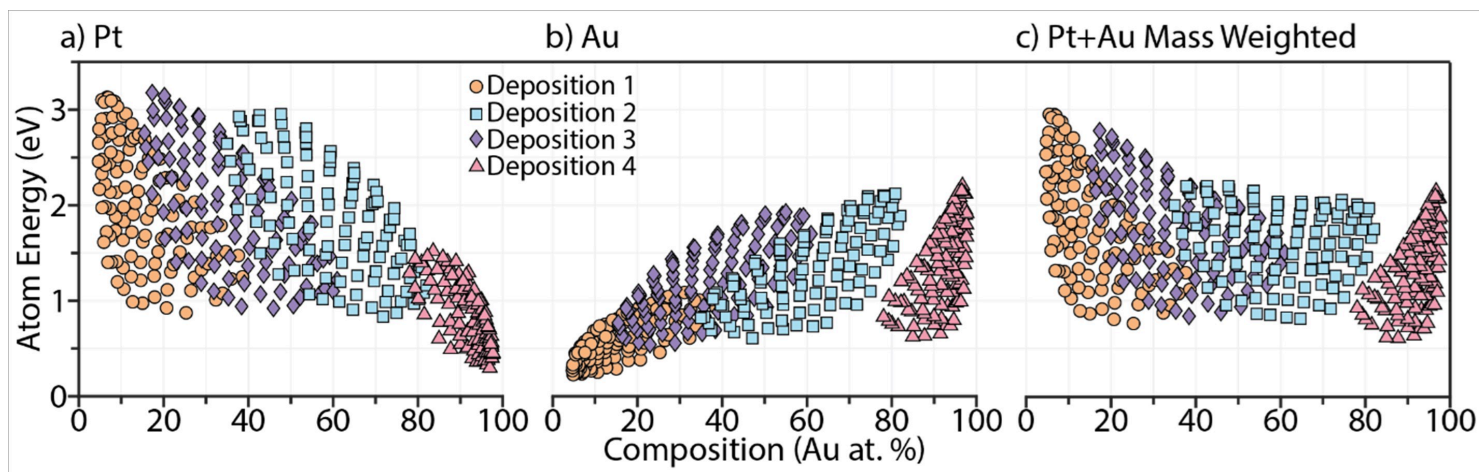


Figure 2

The calculated average kinetic energy of (a) incoming Pt atoms, (b) Au atoms, and (c) a mass-weighted average for all 4 depositions derived from SIMTRA-predicted energy distributions.

Robotically Automated, Paralellized and Integrated Design

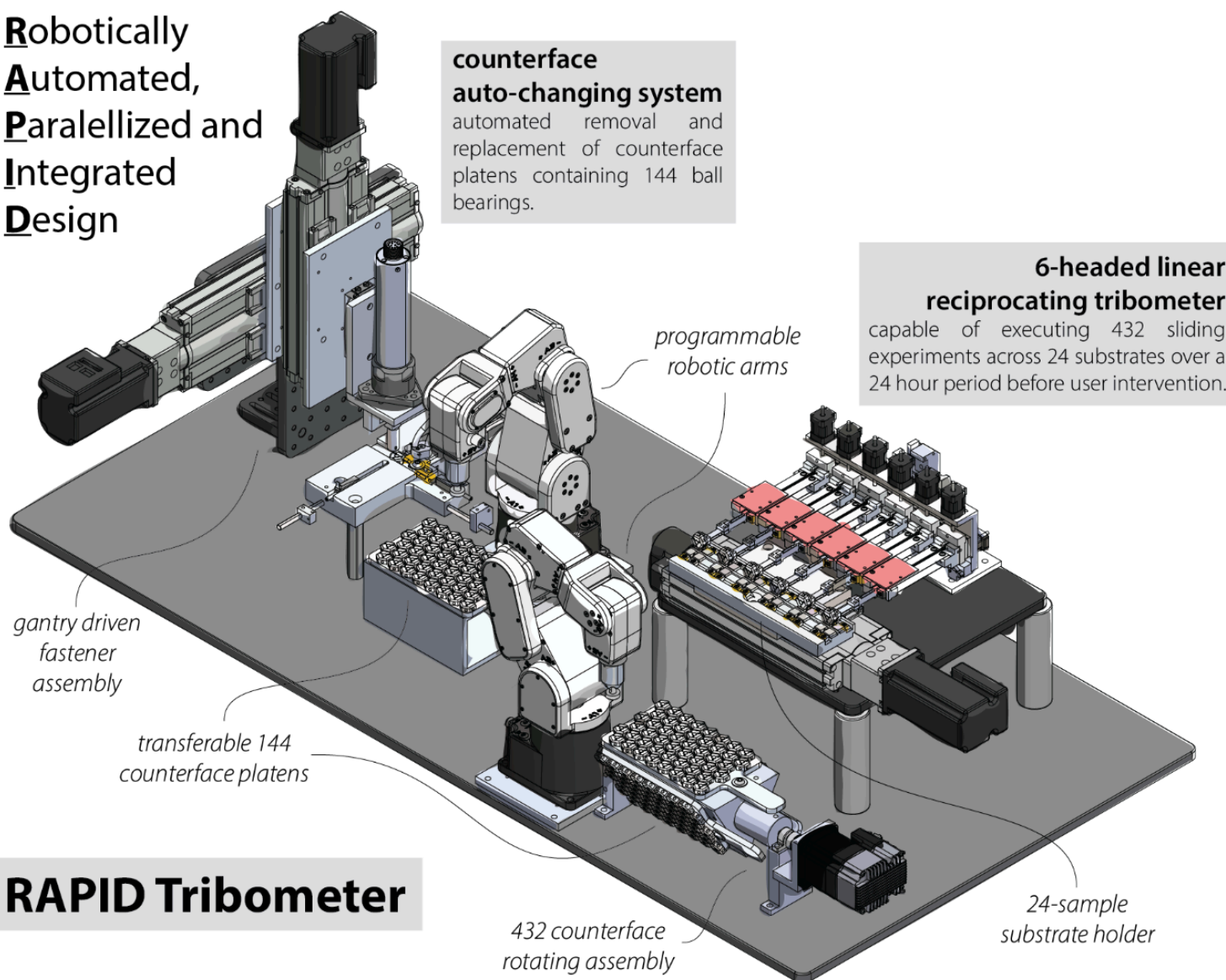


Figure 3

Illustration of the RAPID tribometer using robotic manipulation of the sample heads and automatic loading of ball holders allowing for 432 experiments to be tested on 24 substrates in a 24-hour period.

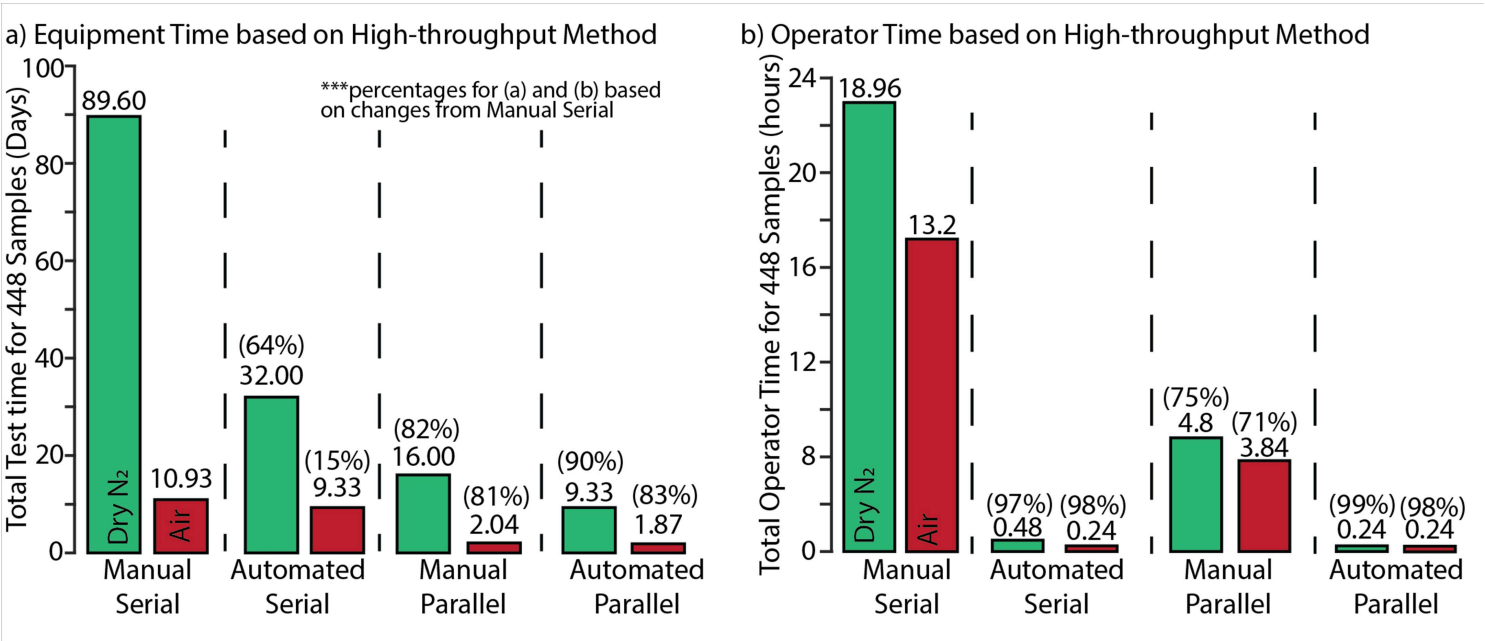


Figure 4

(a) The estimated total time for a tribometer to test 448 samples and (b) the total operator time to complete all 448 samples.

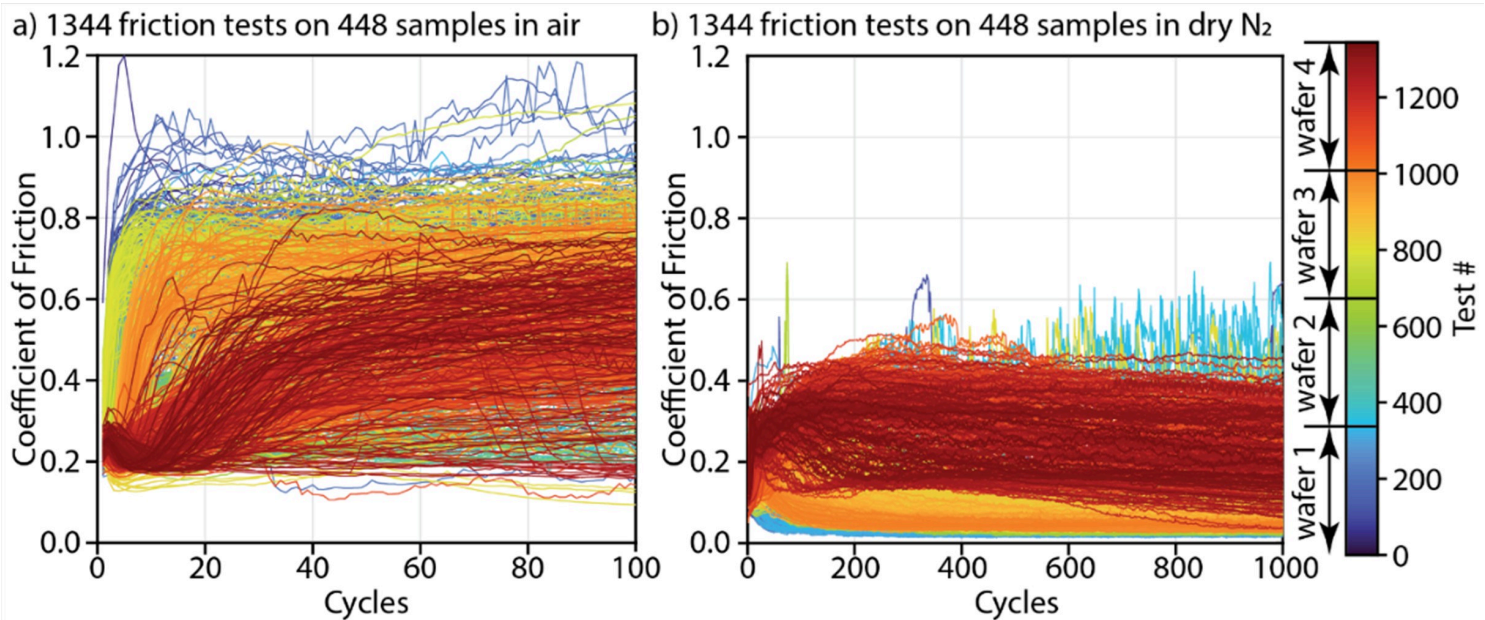


Figure 5

All raw average cycle coefficient of friction data for coatings tested in (a) lab air and (b) dry N₂. A total of 1344 average friction traces were produced for each environment tested.

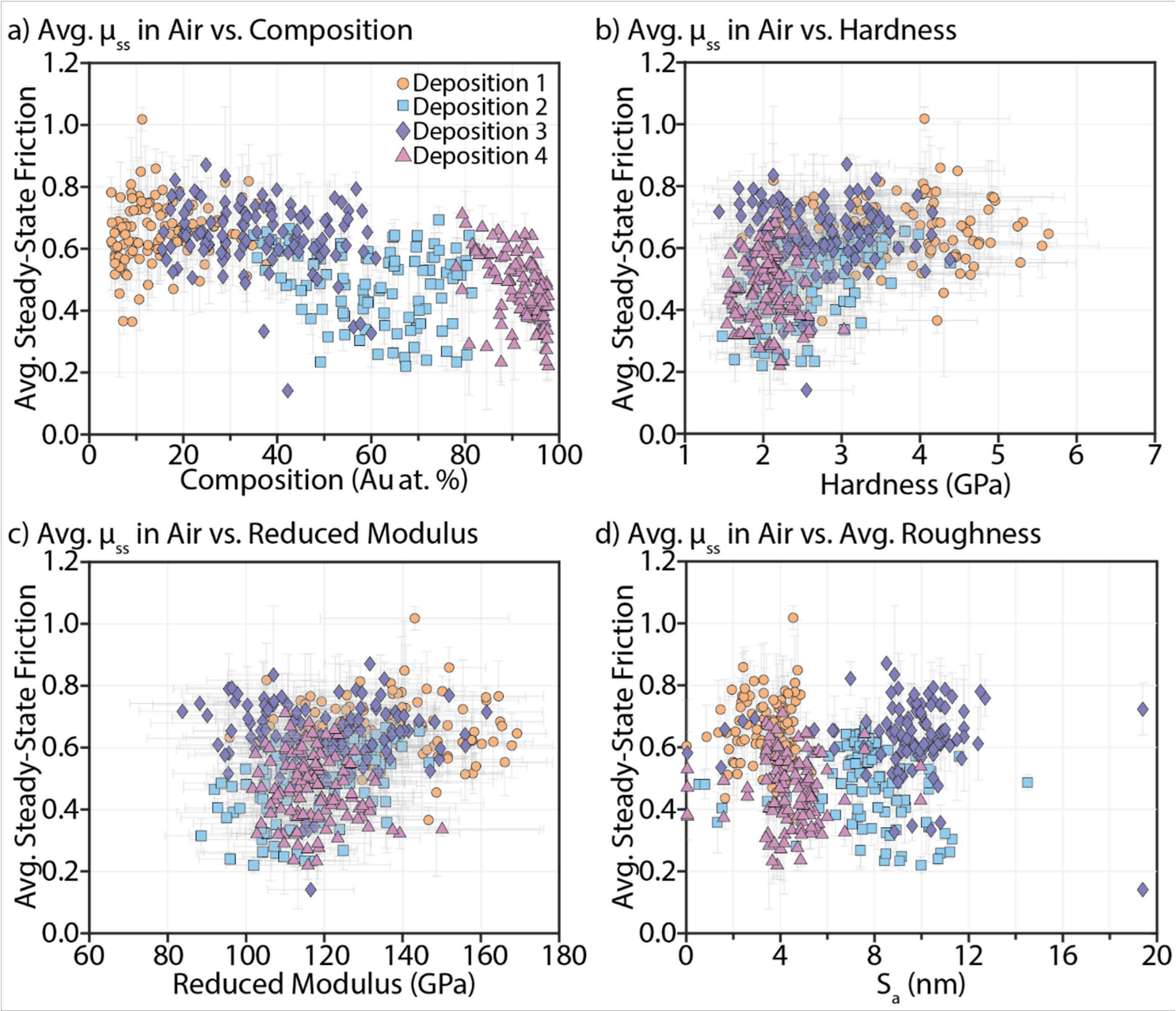


Figure 6

Steady-state coefficients of friction measured in lab air averaged across all three replicates for all 448 samples compared to (a) the coating composition calculated using SIMTRA, (b) the nanoindentation hardness, (c) reduced modulus, and (d) the average roughness (S_a) measured using AFM.

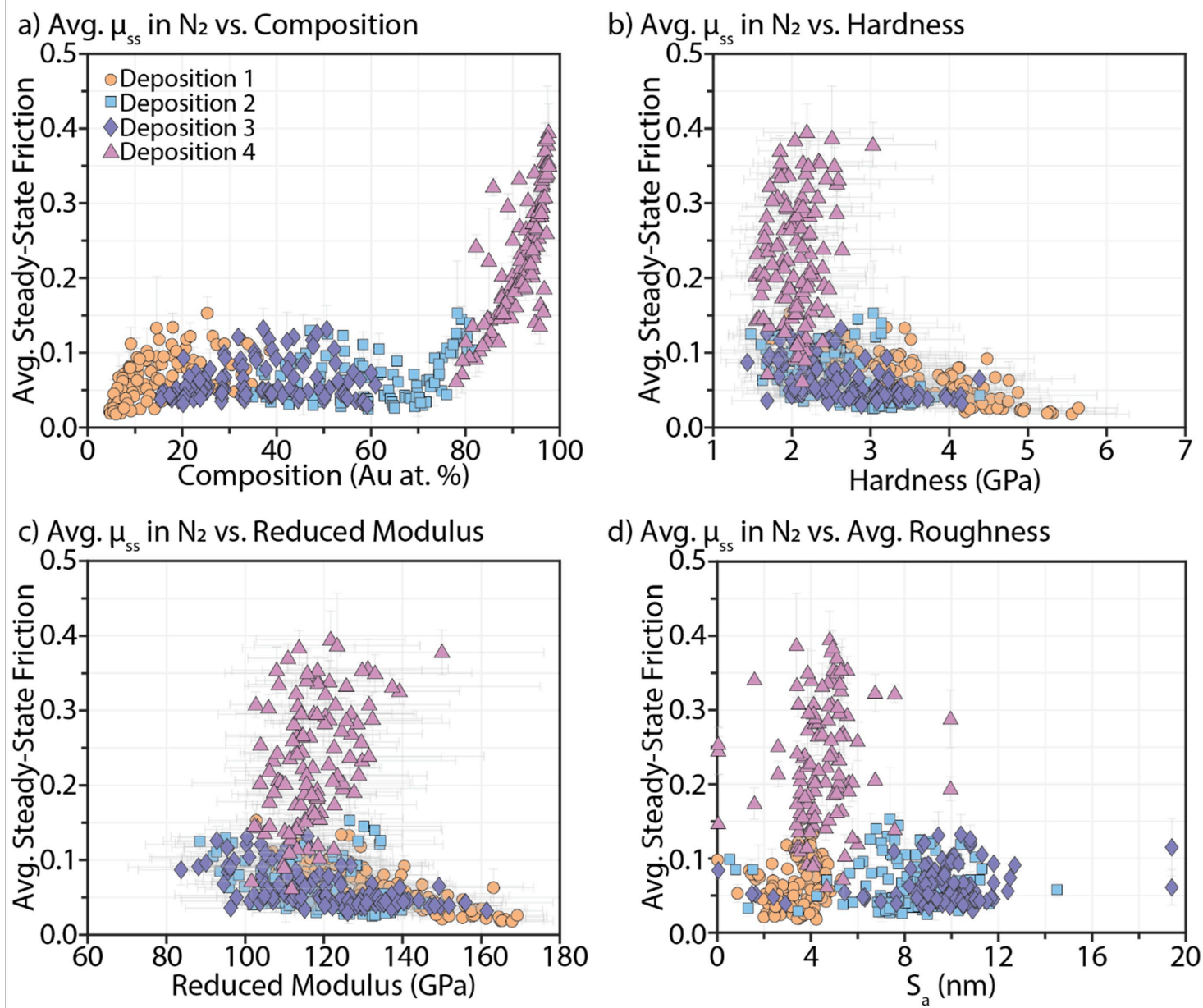


Figure 7

Steady-state coefficients of friction measured in dry N₂ averaged across all three replicates for all 448 samples compared to (a) the coating composition calculated using SIMTRA, (b) the nanoindentation hardness, (c) reduced modulus, and (d) the average roughness (S_a) measured using AFM.

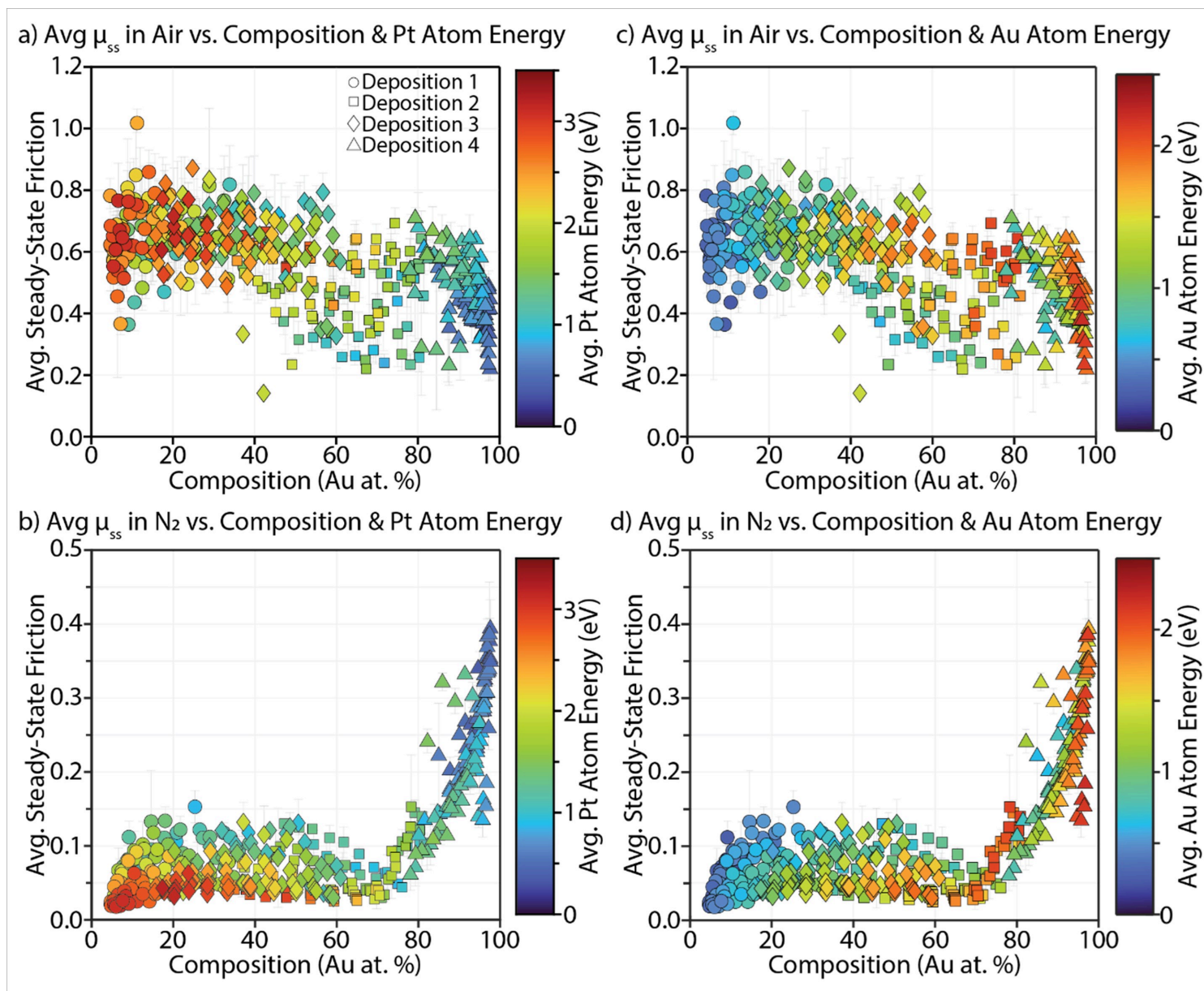


Figure 8

Steady-state coefficients of friction compared to coating composition in (a) lab air and (b) dry N₂ color coded by the calculated average Pt atom energy via SIMTRA and (c) lab air and (d) dry N₂ color coded by the calculated average Au atom energy via SIMTRA.

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