

## Protective and High-temperature Coatings Room Town & Country D - Session MA4-1-TuA

### High Entropy and Other Multi-principal-element Materials I

**Moderators:** Shih-Hsun Chen, National Yang Ming Chiao Tung University (NYCU), Taiwan, Pavel Soucek, Masaryk University, Czechia

#### 1:40pm MA4-1-TuA-1 Phase-Adjustable High-Entropy Alloy Coatings Prepared via Thermal Spray Process, Shih-Hsun Chen [brucechen@nycu.edu.tw], NYCU, Taiwan **INVITED**

The alloy design in HEAs has impacts the resultant microstructure, phase structure, and hardness and wear resistance. Careful selection of HEAs elements is critical depending on the intended application of the alloy. Development of HEAs alloys follows careful selection of elements such as Co, Cr, Ni, Al, Fe, Ti among others. Mechanical properties in HEAs depend on existing phases whether single phase (BCC or FCC) or a mixture of phases and intermetallics or oxides. AlCoCrFeNi alloy has been widely studied and is considered an excellent base for additional strengthening strategies through compositional optimization. In this study,  $Al_{0.5}CoCrFeNi_2Ti$  powders were prepared using the gas atomization method, and annealing treatments were performed to characterize the phase transformation behavior, providing essential insights into the effects of thermal energy during the atmospheric plasma spraying process. To further explore these effects, the study examined effects of plasma energy and powder size in the production of  $Al_{0.5}CoCrFeNi_2Ti$  coatings. The characterization of both powders and coatings was consolidated to better understand the influence of Ti addition on the  $Al_{0.5}CoCrFeNi_2$  HEA system and to assess the performance of its plasma-sprayed coatings.

#### 2:20pm MA4-1-TuA-3 Three Noble Metals, Three Different Stories: Unraveling the Complex Behavior of Cu, Ag, and Au in CrMnFeCoNi High-Entropy Alloy Thin Films, Salah-eddine Benrazouq [salah-eddine.benrazouq@univ-lorraine.fr], Institut Jean Lamour - Université de Lorraine, France; Ekaterina V. Gunina, Svyatoslav Povarov, School of Physics and Engineering, ITMO University, Russian Federation; Jaafar Ghanbaja, Sylvie Migot, Alexandre Nominé, Jean-François Pierson, Valentin A. Milichko, Institut Jean Lamour - Université de Lorraine, France

Advanced electronic devices and sensing technologies demand materials with precisely tunable electrical and optical properties, alongside excellent structural stability. While high-entropy alloys (HEAs) show promise for such applications due to their unique multi-element composition, controlling their functional properties remains challenging. Noble metals (Cu, Ag, Au) were strategically chosen for this study due to their similar electronic configurations and increasing metallic radii ( $r_{Cu}=0.127$  nm,  $r_{Ag}=0.144$  nm,  $r_{Au}=0.147$  nm) compared to the average metallic radius of the Cantor alloy (0.125 nm).

This study unravels the distinct stories of how Cu, Ag, and Au additions transform the structure and properties of CrMnFeCoNi Cantor alloy thin films, revealing behaviors that challenge our initial expectations. Using DC magnetron co-sputtering, we systematically investigated these transformations through comprehensive characterization including X-ray diffraction (XRD), high-resolution transmission electron microscopy (HRTEM), electrical resistivity measurements, and non-linear optical response.

Each noble metal reveals a unique chapter in phase evolution and property modification, far from the expected systematic progression based on atomic size. Copper tells a story of structural preservation, maintaining the fcc structure while systematically reducing nano-twin density, culminating in near-zero TCR (-2.86 ppm/K) at 37 at% Cu. Silver writes a different narrative through unexpected phase separation, creating artistic patterns of nano-precipitates with a characteristic tweed-like microstructure, despite predictions of solid solution formation. Gold presents perhaps the most surprising tale, where HRTEM unveils large grains with intricate twin boundary networks, contrasting sharply with its apparent amorphous nature in XRD analysis and defying expectations based on atomic size considerations.

These distinct structural modifications yield equally diverse functional properties. Cu-modified films demonstrate precise control over electrical behavior while maintaining metallic characteristics, aligning with initial predictions. Ag-modified films combine decreased electrical resistivity with enhanced second harmonic-generation (SHG) response, providing unexpected opportunities for multifunctional properties. Au-modified films exhibit unique optical properties tied to their complex grain structure.

These three different stories highlight how noble metals can orchestrate dramatically different transformations in HEA thin films, while comparing their properties with sustainability metrics offers guidance for future technological implementations

#### 2:40pm MA4-1-TuA-4 Mechanical, Tribological and Corrosion Behavior of CoCrFeNiMn High-Entropy Thin Films, Lin Wu [lin.wu2@mail.mcgill.ca], McGill University, Canada; León Zendejas Medina, McGill University, KTH Royal Institute of Technology, Canada; Richard Chromik, Janine Mauzeroll, McGill University, Canada

The CoCrFeNiMn (Cantor alloy) thin films deposited under ambient and high temperature conditions have been studied from the aspect of microstructure, mechanical, tribological and corrosion properties. The Cantor films were deposited by pulsed direct current magnetron sputtering on silicon wafers at ambient and 350 °C. An FCC phase appeared in both ambient and high temperature films, with small amounts of an unidentified secondary phase. Using nanoindentation, film hardness (H) and reduced elastic modulus (Er) were measured.

Micro-tribology testing was conducted using a 20  $\mu$ m radius spherical diamond tip in the dry air atmosphere (RH% value under 4.0%), applying normal loads ranging from 2 to 7 mN. The worn surfaces were characterized by atomic force microscopy. Schiffrin's model was used to evaluate the elastic and plastic components of the friction. The corrosion behaviors of the films were studied by using anodic polarization in 3.5% NaCl, followed by a tribological testing carried out on the corroded surfaces, to address the correlation of corrosion response with the phase composition, mechanical properties and tribological behavior of the films.

#### 3:00pm MA4-1-TuA-5 Microstructure and Mechanical Properties Evaluation of CoCrNiTiAl Multiple-Principal Element Alloy Thin Films: Effect of TiAl Additions, Pongpak Chiyasak [pongpak.c@ku.th],

Department of Materials Engineering, Faculty of Engineering, Kasetsart University, Thailand; Jun-Xing Wang, Ming Chi University of Technology, Taiwan; Chia-Lin Li, Center for Plasma and Thin Film Technologies, Ming Chi University of Technology, Taiwan; Surapit Posri, Thanawat Santawee, Worawat Wattanathana, Aphichart Rodchanarowan, Department of Materials Engineering, Faculty of Engineering, Kasetsart University, Thailand; Jyh-Wei Lee, Ming Chi University of Technology, Taiwan

Multiple-principal element alloy thin films have attracted lots of interest from academia due to their optimized properties for both functional and structural applications, for example, high strength, good thermal stability, cost efficiency, and lower stacking fault energy. Several previous studies have shown that adding Ti and Al as alloying elements into CoCrNiTiAl multiple-principal element alloy thin films could further enhance their performance. In general, Ti element can transform the face-centered cubic structure into the amorphous structure, while Al generates body-centered cubic (BCC) structures, promoting higher hardness and wear resistance. However, the effect of the addition of Ti and Al elements into CoCrNiTiAl multiple-principal element alloy thin films is still unexplored.

In this work, the CoCrNiTiAl multiple-principal element alloy thin films with different amounts of Ti and Al contents were fabricated by the co-sputtering of TiAl and CoCrNi targets through a hybrid magnetron sputtering system. The CoCrNi and TiAl target were connected with a high power impulse magnetron sputtering system and a radio frequency power supply, respectively. The power input of the TiAl target was adjusted to achieve multiple-principal element alloy thin films with different TiAl contents. The field-emission (FE) scanning electron microscopy, FE-electron probe microanalyzer, X-ray diffraction, nanoindentation, tribometer, and electrochemical workstation were used to characterize microstructure, chemical composition, phase evolution, hardness, wear resistance, and corrosion behavior of multiple-principal element alloy thin films. The effect of TiAl contents on the microstructure and mechanical properties of CoCrNiTiAl multiple-principal element alloy thin films were discussed in this work.

**Keywords:** multiple-principal element alloy, CoCrNiTiAl, phase transformation, mechanical properties

# Tuesday Afternoon, May 13, 2025

4:00pm **MA4-1-TuA-8 Effects of Deposition Parameters and Post-Annealing Treatment on the Microstructure and Mechanical Properties of TiZrNbTaMo High Entropy Alloy Films**, *Chia-Lin Li [chialinli@mail.mcut.edu.tw]*, Center for Plasma and Thin Film Technologies, Ming Chi University of Technology, Taiwan; *Sen-You Hou*, Department of Materials Science and Engineering, National Tsing Hua University, Taiwan; *Li-Chun Chang*, Ming Chi University of Technology, Taiwan; *Bih-Show Lou*, Chemistry Division, Center for General Education, Chang Gung University, Taiwan; *Jyh-Wei Lee*, Department of Materials Engineering, Ming Chi University of Technology, Taiwan; *Po-Yu Chen*, Department of Materials Science and Engineering, National Tsing Hua University, Taiwan

TiZrNbTaMo high-entropy alloys (HEAs) with a body-centered cubic structure are known for their excellent compressive yield strength and significant compressive plasticity, retained even in thin film forms. These properties make them promising for various applications. The deposition parameters significantly influence the density and microstructure of thin films, affecting mechanical properties. In this study, TiZrNbTaMo high-entropy alloy films (HEAFs) were prepared using high power impulse magnetron sputtering (HIPIMS), DC, and RF power sources. The effects of pulse frequency and duty cycle in HIPIMS on their structure and properties were systematically investigated. The microstructure and crystal structure of TiZrNbTaMo HEAFs were characterized using transmission electron microscopy (TEM) and X-ray diffractometry (XRD), while their mechanical properties were evaluated by nanoindentation. TiZrNbTaMo HEAFs deposited by HIPIMS exhibited increased hardness due to higher peak power density resulting in the coexistence of amorphous and nanocrystalline structures. However, the highest hardness of 5.78 GPa was achieved with RF power source, which was attributed to more nanocrystalline content and stacking faults. To further improve the mechanical properties of HEAs, post-annealing is used to modify the grain size and structure by inducing microstructures such as stacking faults and twins. In this study, the effects of post-annealing at 900 °C on microstructure and mechanical properties were investigated, and detailed features such as grain sizes, annealing twins, and variations in mechanical properties were discussed.

4:20pm **MA4-1-TuA-9 High Entropy Alloys Coatings for Inertial Confinement Fusion Hohlräume**, *Leonardus Bimo Bayu Aji [bayuaji1@llnl.gov]*, *Daniel Goodelman*, *David Strozzi*, *Brandon Bocklund*, *Scott Peters*, *Alison Engwall*, *Swanee Shin*, *Gregory Taylor*, *Eunjeong Kim*, *James Merlo*, *Sergei Kucheyev*, Lawrence Livermore National Laboratory, USA

Hohlräume, centimeter-scale spherocylindrical heavy-metal cans with wall thickness of 10 - 100  $\mu\text{m}$ , are a key component of an indirect-drive inertial confinement fusion (ICF) target as it determines the x-ray drive that implodes the fuel capsule. A previous study [Jones et al., Phys. Plasmas 14, 056311 (2007)] has demonstrated the feasibility of improving the x-ray drive by using a hohlraum made from a mixture ("cocktail") of elements, instead of a single element hohlraum, such as Au or U traditionally used for ICF. Here, we present our results on developing sputter-deposited heavy-metal high-entropy alloys (HEAs) for hohlraums with improved x-ray drive, high electrical resistivity to support a magnetized ICF, and material properties that are compatible with the ICF target fabrication process.

This work was performed under the auspices of the U.S. DOE by LLNL under Contract DE-AC52-07NA27344 and LDRD project 23-ERD-005.

4:40pm **MA4-1-TuA-10 Evaluation of the Microstructure and Electrocatalytic Performance of FeNiMoWCu<sub>x</sub> High-entropy Alloy Thin Film**, *Kuan-Chen Lin [love17321125@gmail.com]*, Ming Chi University of Technology, Taiwan; *Bih-Show Lou*, Chang Gung University, Taoyuan city, Taiwan; *Chia Lin Li*, *Naveen Karuppusamy*, *Thi Cam Tuyen*, *Jyh-Wei Lee*, Ming Chi University of Technology, Taiwan

Since Professor Yeh's introduction of high-entropy alloys (HEAs) in 2004, research on HEA materials has garnered worldwide attention due to their exceptional properties. Certain HEA materials have demonstrated great potential as electrocatalysts, attributed to their unique structures and compositions. In this study, FeNiMoWCu<sub>x</sub> HEA thin films with varying Cu contents were synthesized on Si wafers and nickel foams using magnetron sputtering. Co-sputtering was performed with equimolar FeNiMoW and pure Cu targets to produce FeNiMoWCu<sub>x</sub> films with  $x = 0, 0.5, \text{ and } 1.0$ . An inclined deposition technique was employed to increase surface roughness and create more active sites on the HEA films. A comprehensive evaluation of the films was conducted, examining their composition, microstructure, crystallographic structure, surface roughness, adhesion, and corrosion

resistance in 0.5 M sulfuric acid. Furthermore, electrochemical oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) performance along with long term stability were carried out using a three-electrode electrochemical configuration in 1.0 M KOH and 1.0 M KOH + 3.5 wt.% NaCl solutions, respectively, to assess their electrocatalytic properties. The findings revealed that the FeNiMoWCu<sub>x</sub> HEA films hold considerable promise for electrode surface modification in water-splitting applications, owing to their improved catalytic activity, structural stability, and excellent corrosion resistance.

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