Advanced Thin Film High Entropy Alloys with Tailorable Microstructure and Mechanical Properties

Davide Vacirca*†[1], Francesco Bignoli[1,2], Andrea Li Bassi[2], James Paul Best[3], Gerhard Dehm[3], Philippe Djemia[1], and Matteo Ghidelli[1]

[1]Laboratoire des Sciences des Procédés et des Matériaux – CNRS – France
[2]Dipartimento di Energia [Milano] – Italy
[3]Max-Planck-Institute for sustainable materials – Germany

Abstract

High entropy alloy thin films (HEAs-TF) have recently gained attention due to their possibility to combine mutually exclusive properties, such as high yield strength, ductility and thermal stability, while offering a wide range of chemical compositions. For this reason, HEAs-TF have potential applications for microscale electrical devices or high-performance coatings (1). However, microstructural control for advanced HEAs-TF (nanocrystalline films, nanolaminates) is often limited by traditional thin film deposition techniques such as magnetron sputtering, hindering the formation of HEAs-TF with large mechanical properties (yield strength, ductility). In this regard, pulsed laser deposition (PLD) was used to prepare advanced HEAs-TF allowing to easily control the nanoarchitecture of nanocrystalline films and ultrafine nanolaminates.

In this context, firstly I will focus on the synthesis of nanocrystalline CoCrCuFeNi HEA-TFs by PLD with a wide range of microstructures (grain size and morphology) obtained simply by controlling the background gas pressure, while comparing with sputter-deposited counterparts. A transition from compact to nanogranular morphology was observed for deposition pressures > 1 Pa, leading to a decrease in mass density and grain size (6.81 g/cm3 and 11 nm, respectively) compared to the compact counterpart (7.72 g/cm3 and 40 nm, respectively). These films by PLD show increased hardness ($\approx$11 GPa) compared to sputter-deposited films ($\approx$8.3 GPa) due to their ultrafine grain structure leading to Hall-Petch strengthening. Similarly, micropillar compression tests show high yield strength of 1.9 GPa with high plastic deformability (30% strain). Post-thermal annealing treatments reveal grain coarsening and segregation of a secondary BCC phase starting at 400$\circ$C for both compact and nanogranular films. However, nanogranular films maintain a lower grain size during annealing ($<$ 20nm), thus retaining higher hardness of 8.5 GPa (Hall-Petch strengthening).

Secondly, I will focus on Al/HEA nanolaminates (NLs) by PLD and magnetron sputtering, aiming to block the propagation of dislocations by controlling the interface density to improve hardness and yield strength (2). Nanolaminates with different atomic compositions have been fabricated by magnetron sputtering, involving semi-coherent (FCC/FCC Al/CoCrCuFeNi) and incoherent (FCC/BCC Al/Al25(CoCrCuFeNi)75) interfaces to further hinder cross-layer dislocation propagation and boost mechanical properties. Among the main results, *in situ* micropillar compression test shows that incoherent interfaces have

---

*Speaker

†Corresponding author: davide.vacirca@lspm.cnrs.fr

greater capability to improve yield strength (up to 2.2 GPa), while still showing great plastic deformability with no appearance of cracks even at 30% deformation. Using PLD, a wide range of bilayer periods () was explored, allowing to prepare ultrafine NLs with a bilayer period as low as 2.5 nm up until 400 nm. These NLs show high hardness, with a maximum of 9.8 GPa for = 25 nm due to the high interface density, despite having a 50% volume fraction of Al.

Our results show how PLD and sputtering can be used to control the nano-engineering of TF-HEAs, resulting in improved and tunable mechanical properties with key implications for industry applications.

(1) N. I. M. Nadzri *et al.* ‘High-Entropy Alloy for Thin Film Application: A Review’, *Coatings*, 12, 2022
(2) A. Sàenz-Trevizo *et al.*, Nanomaterials by design: a review of nanoscale metallic multilayers Nanotechnology, 31, 2020