CoCrNi and FexCoCrNi(100-x) complex compositional alloy thin films: Defect-induced microstructural evolution and microscale mechanical behavior

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Abstract

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Complex compositional alloys (CCAs) are recent materials characterised by a high number of atomic constituents, which are gaining interest due to the possibility of finely tuning their mechanical behaviour by adjusting their composition, potentially leading to large strength/ductility balance, especially for FCC nanocrystalline alloys (1). From the CoCrFeMnNi "Cantor" alloy, one of the frequently studied ternary compositions is CoCrNi, which reported hardness (H) and elastic modulus (E) values up to 9 and 180 GPa, respectively, due to a columnar structure with planar defects (2). The addition of Fe to form FeCoCrNi stabilised the FCC phase with a small grain size of ~20 nm, affecting H mildly up to ~10 GPa (3). These studies, however, were mainly conducted on bulk alloys, and research on complex compositional alloys thin films (CCAs-TF) is still in its early stages, particularly concerning the relationship between compositional variation, microstructural evolution, and their effect on mechanical behaviour, requiring the need of advanced characterisation techniques capable of probing the mechanical behaviour of sub-micrometre scale materials. Moreover, a nano-engineered approach to developing new multilayered thin film nanoarchitectures consisting of alternating layers and phase structures with boosted mechanical properties remains an open field.

Here, we synthesised CoCrNi and Fex(CoCrNi)100-x CCA-TFs by magnetron sputtering, in which the thicknesses were controlled from 15 nm up to 730 nm, and the Fe content was varied from 13 up to 29 % at. For each of the four compositions, using HRTEM, we investigated the evolution of the microstructure and phase structure as the thickness increased. With the help of nanoindentation (CSM), we analysed how the increasing Fe content affected the mechanical behaviour. Thick CoCrNi film shows a columnar structure with an average

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grain size of _~26 nm and a (111) textured FCC structure with a minor HCP (100) phase segregation. Additionally, we observed a transition from HCP to co-existing HCP/FCC upon increasing thickness from 15 to 40 nm. The presence of the minor HCP structure was attributed to a high density of Intrinsic Stacking Faults (ISF) and twinning within the FCC structure, which was responsible for a high H = 9.7 GPa and E=204.4 GPa as reported by nanoindentation and optoacoustic techniques.

With the addition of Fe, we retained the columnar structure while stabilising the FCC phase, reporting a preferred (111) orientation without phase segregation due to good miscibility of Fe within CoCrNi. With no segregation of Fe, and fewer defects like stacking faults owing to a higher stacking fault energy 32.5 mJ m–2 (4), H and E decrease down to 7.6 and 185 GPa.

While the FCC phase was stabilised in FeCoCrNi, individual layers of CoCrNi and Fe can lead to different phases and potential Fe segregation to obtain strong interfaces. For this reason, we synthesised _~1.4 μm thick CoCrNi/Fe (FCC/BCC) nanolaminates with bi-layer periods () of 70 and 35 nm, featuring incoherent interfaces aimed at blocking dislocations and crack propagation. We show high adhesion at the interfaces with limited crack formation during indentation, while compression of micropillars resulted in high yield strengths (2.5 GPa) without fracture up to 12%. Owing to a high density of interfaces and thickness confinement, a high plasticity of up to 40% was obtained.

Overall, our study shed light on the microstructure–mechanical properties of CoCrNi, Fex(CoCrNi)100-x thin films, and CoCrNi/Fe nanolaminates, with potential applications in hard coatings and microelectronics.

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