Materials Science and Engineering Dissertation Defense

Probing High-Temperature Oxidation Mechanisms of Nanocrystalline Ternary Alloys

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Abstract

High-temperature oxidation of nanocrystalline (NC) alloys has garnered widespread research interest due to their appreciable thermomechanical properties, attributed to grain refinement in NC alloys. Immiscible microstructurally-stable binary NC Cu-Ta alloys offer exceptional potential for high-temperature applications where a combination of strength, hardness, and oxidation resistance is crucial. Particularly, Cu-based alloys provide high ductility while immiscible Ta imparts high-temperature mechanical strength in the form of uniformly dispersed nanoclusters within the microstructure. The incorporation of oxygen-active elements such as Cr, particularly as a “third-element” (or second solute) in binary alloys, has been established as the ideal choice of alloying element to impart enhanced oxidation resistance in the binary NC Cu-Ta system, owing to the accelerated and selective formation of stable, protective Cr2O3 scales. However, the incorporation of Cr inherently modifies Ta segregation tendency in immiscible Cu-Ta alloys, thereby impacting ternary microstructures and modifying secondary phase precipitation in ternary alloy systems. The addition of Cr also plays a significant role in dictating thermodynamic driving forces (such as free energy of oxide formation, alloy stoichiometry and microstructure, and alloy grain size) as well as kinetic influences (such as the diffusion coefficients of participating ionic species and diffusion conditions) involved during high-temperature oxidation reactions. This work explores the concomitant effect of nanocrystallization and solute engineering via Cr solute addition in binary NC Cu-Ta alloys, to systematically study the role of grain size and alloy composition on impacting the high-temperature oxidation of ternary NC Cu-Ta-Cr alloys. The addition of increasing concentrations of Cr allows the ternary Cu-Ta-Cr alloys to effectively transition from low-solute high-oxidation prone alloys to high-solute low-oxidation alloys. Proposed oxidation mechanisms in low-solute and high-solute conditions are presented by considering fundamental thermodynamic and kinetic concepts. This dissertation utilizes advanced materials characterization techniques such as scanning transmission electron microscopy (STEM), electron dispersive spectroscopy (EDS), in-situ oxidation using powder x-ray diffraction (P-XRD), and thermogravimetric analysis (TGA), to study the onset and progression of high-temperature oxidation in nanocrystalline Cu-Ta-Cr ternary alloys. Overall, this research establishes a framework for the design of alloys exhibiting superior thermomechanical stability and enhanced oxidation resistance for prolonged durability in advanced engineering applications.

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