ABSTRACT

The present work leverages atomistic simulation techniques to investigate the influence of site-specific solute segregation and grain boundary (GB) structural transition on diffusion behaviour, radiation tolerance and mechanical response in Ni-Nb alloys. In addition, data driven approaches, such as machine learning methods, are employed to predict the segregation energy of Nb solute in both bicrystal and nanocrystalline (NC) Ni GBs. To obtain the equilibrium structure of the Ni-Nb binary alloys at various temperatures and dopant concentrations, hybrid Monte Carlo/molecular dynamics simulations (MC/MD) are performed on both bicrystal and NC specimens. Initially, the role of Nb segregation on GB structural transformations and mechanical response is investigated in a bicrystal Σ5(310) Ni–Nb system. The simulation results indicate that at lower dopant concentrations, Nb atoms preferably segregated at the tip of the kite structure. With an increase in dopant concentration and temperature, the Nb atoms segregated at other sites in the GB as well as into the matrix, leading to disordering of the GB structure. Subsequently, the effect of Nb doping on the mechanical behaviour has been investigated using simulated tensile tests in the bicrystal Ni–Nb alloy. The stress-strain curve of the Ni-Nb system at 300 K revealed an increase in the strength and ductility with the dopant concentration up to a certain limit (0.4 at. % Nb) and decreased thereafter. The superior mechanical response of the Ni-0.4Nb system is attributed to the formation of stacking faults and sessile dislocations during deformation. Further, the effect of segregation induced structural transition on GB diffusivity is studied in same bicrystal Σ5(310) GB across various Ni-Nb alloy compositions (0.5, 1.0, 1.5, 2.0, 2.5, and 5.0 at. % Nb) and at different temperatures (1200 K, 1300 K, 1400 K, and 1500 K). The simulation results show that the GB diffusivities of solute (Nb) and solvent (Ni) atoms exhibit distinct composition/temperature dependencies. Furthermore, the diffusivity of Ni atom decreases in the Ni-0.5Nb specimen compared to the pure Ni at lower temperatures (1200 K and 1300 K) due to preferential segregation of Nb atoms at the most favorable sites. Consequently, Nb atoms exhibit reluctance to migrate to other sites even under a driving force, and the overall diffusion process slows down. However, as the solute concentration and temperature increase, the GB structures become disordered, altering the GB diffusion mechanism. Afterward, the spectral nature of GB atoms is explored in NC Ni, and found that the GBs have a skew-normal distribution for excess atomic volume and excess atomic energy. Furthermore, comprehensive segregation energy calculations are performed for each GB site, revealing the preferential affinity of Nb towards GB sites in NC Ni. Moreover, simulations conducted at higher temperatures (i.e., 1200 K) and higher dopant concentrations in NC Ni reveal that not all GBs within the microstructure transform into amorphous complexions following Nb segregation; instead, some GBs remain ordered. The simulated tensile tests conducted at 500 K and 1200 K reveal that the strengthening in NC Ni-Nb enhances with the increase in dopant concentration. Additionally, the simulated shear test conducted at 1200 K shows that the deformation in NC Ni-5Nb occurs more uniformly than pure NC Ni due to formation of

thicker amorphous GBs in the former one. Furthermore, atomistic simulations are performed to investigate the role of amorphous grain boundaries (AGBs) in mitigating radiation damage and influencing the mechanical properties of the Ni-Nb alloy. In this regard, AGBs of various GB thickness (i.e., 1 nm, 2 nm, 3 nm, and 4 nm) have been constructed, and the radiation induced damage study are performed using primary-knock-on atom (PKA) with energies of 2.5 keV, 5 keV, and 10 keV, respectively, at 300 K. The simulation results indicate that as the thickness of AGBs increases from 1 nm to 4 nm, a significant reduction in the defect region width is observed during both the peak damage and residual stages. Notably, for an AGB thickness of 4 nm, the defect width decreases to one-fourth of that in the thin ordered pure Ni GB, with defects being completely absent in the bulk region under the simulated conditions**.** Additionally, simulated tensile tests were conducted to evaluate the impact of irradiation damage on the mechanical properties of different GBs. The simulated tensile results reveal that the thinner AGBs (1–2 nm) retained more strength after irradiation than the thicker ones (3–4 nm). This contrasting behavior is primarily attributed to the greater number of irradiation-induced defects generated in the bulk region of thinner AGBs (1–2 nm), which contribute to radiation-induced strengthening. In contrast, in thicker AGBs, these defects are more effectively absorbed, thus diminishing their strengthening effect. Finally, several machine learning approaches, namely linear regression models, decision trees-based models, and a deep learning-based model are utilized to predict the segregation energy of Nb solute in both bicrystal and NC Ni GBs. In this regard, per-atom metrics (e.g., local stress, Voronoi volume, coordination number, etc.) are extracted from undecorated pure Ni GBs. The results show that XGBoost consistently achieves the highest prediction accuracy, and the performance gap relative to other models (e.g., Random Forest, Extra Trees, Artificial Neural Networks) becomes more pronounced in nanocrystalline specimen with greater structural diversity.

Keywords: Amorphous boundaries; Atomistic simulations; Complexion; Diffusion; Dislocation; Grain Boundaries; Machine learning; Mechanical behaviour; Nanocrystalline; Ni-Nb alloy; Primary-knock-on atom; Radiation damage; Radiation-induced strengthening; Segregation; Structural transition; XGBoost.