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Paper Title: Effects of Gd on the microstructure and mechanical properties of GdxCoCrFeNiV0.4 high-entropy alloys
Content :

The concept of high-entropy alloys was initially proposed at the end of the twentieth century, marking a significant advancement in alloy design [1,2]. These alloys consist of five or more primary elements and show great potential in structural applications due to their superior mechanical properties, wear resistance, corrosion resistance, and high strength [[3], [4], [5], [6], [7]]. Current research on high-entropy alloys has primarily focused on Cantor alloys, composed mainly of 3d transition metals, Senkov alloys, which primarily consist of refractory metals, and low density high-entropy alloys containing light elements such as Al and Mg. The majority of studies in this field have centered on the formation of mono-phase structures [[8], [9], [10]]. One notable variant, the CoCrFeNi high-entropy alloy, has emerged as a relatively well-studied alloy system. This alloy exhibits superior tensile ductility and fracture toughness at room temperature, although it does have relatively low strength [11,12].

In order to optimize CoCrFeNi high-entropy alloys, researchers from various countries have conducted extensive studies. These studies aim to enhance the properties of the alloys by incorporating additional alloying elements such as Mn [13,14], Al [[15], [16], [17]], Ti [[18], [19], [20]], Cu [21,22], etc. CoCrFeNiMn high-entropy alloys have a single FCC phase, and they have a good toughness. Salishchev et al. [23] prepared CoCrFeNiMn by vacuum arc melting and the alloy had an average hardness of 170 HV, a tensile strength of 491 MPa, and a plastic strain of 71%. Li et al. [24] prepared Al_xCoCrFeNi alloys using the arc melting plus casting method and demonstrated that the presence of Al promotes the formation of a BCC matrix. They also observed that an increase in Al content leads to lattice distortion and enhances the alloy. The hardness of Al_3CoCrFeNi alloys was measured to be as high as 740 HV. Shun et al. [25] studied the impact of Ti addition on the microstructure and mechanical properties of $CoCrFeNiTi_x$ alloys. They identified the presence of (Ni,Ti)-rich R-phase and (Cr,Fe)-rich σ-phase in $CoCrFeNiTi_{0.3}$ high-entropy alloy, while (Ti,Co)-rich Laves phase and R+σ-phase were found in $CoCrFeNiTi_{0.5}$. The authors concluded that the solid solution strengthening of the FCC matrix and the formation of a hard secondary phase were the key factors influencing the enhancement of the alloy. In another study [26], researchers utilized friction stir processing (FPS) to produce a CoCrFeNiCu alloy. Two distinct phases, a Cu-poor FCC1 phase and a Cu-rich FCC2 phase, were identified in the alloy. The resulting CoCrFeNiCu high-entropy alloy exhibited a hardness exceeding 380 HV and a yield strength exceeding 1150 MPa. Apart from the mentioned alloying elements, the addition of V element was noted for its high temperature stability, aiding in grain

refinement, solid solution strengthening, and reducing Cr element segregation within the alloy. Research has indicated that in CoCrFeNiV$_x$ alloys, with x values below 0.7, enhancements in hardness and tensile strength of the high-entropy alloy can be achieved without altering the phase structure of CoCrFeNi alloy. Conversely, for x values exceeding 0.7, the presence of a σ phase with a square crystal structure was observed in the alloy, with the peak strength of this phase increasing proportionally to the V content [27,28]. Zhang et al. [29] prepared CoCrFeNiV through vacuum hot-pressing sintering, where the iso-atomic inclusion of the V element led to a transformation in the microstructure of the alloy from the FCC phase to a combination of FCC phase and σ phase. The σ phase, known for its hardness and brittleness, significantly enhanced the strength and hardness of the alloy, resulting in brittle fracture with a minor amount of ductile fracture.

Rare earth elements possess distinctive electronic structures and physicochemical properties. Incorporating rare earth elements has been shown to enhance the formation of intermetallic phases and significantly enhance the characteristics of high-entropy alloys [[30], [31], [32], [33]]. Specifically, the rare earth element Gd has a propensity to form a stable hexagonal structure (HS) when combined with elements like Co and Ni due to its low negative mixing enthalpy and significant radius difference. Recently, Gd has been increasingly utilized in strengthening CoCrFeNi high-entropy alloys. For example, in Zhang's study [34], the impact of Gd content on CoCrCuFeNi alloys was examined. It was discovered that Gd altered the phase structure of the original alloys, resulting in the formation of a new Cu-rich hexagonal structure within the face-centered cubic matrix. The hardness was significantly enhanced with increasing Gd content. Specifically, the hardness of Gd increased proportionally with the rise in Gd content. The yield strength of the Gd$_{0.3}$CoCrCuFeNi alloy peaked at 1132 MPa, while the fracture strength reached 1836 MPa. Long et al. [35] conducted a study on the impact of Gd elemental content on the structure and mechanical properties of CoCrFeNi. Their findings revealed that the addition of Gd transformed the alloy from a FCC structure to one consisting of a FCC matrix, a GdNi$_5$-type HS phase, and a small quantity of Gd-rich oxide nanoparticles. With the presence of Gd-rich oxide nanoparticles, the tensile yield strength of the Gd$_{0.05}$CoCrFeNi alloy notably rose from 546 MPa to 859 MPa, and the ultimate tensile strength increased from 733 MPa to 991 MPa. Despite a slight decrease in elongation from 20.8% to 19.4%, plasticity was largely preserved. The plasticity of the alloy is primarily influenced by the even dispersion of the GdNi5-type HS phase within the matrix, which facilitates dislocation slip.

This study focuses on the selection of CoCrFeNiV$_{0.4}$ as the base alloy to prevent plasticity degradation through phase structure modification and enhance mechanical properties by incorporating the rare earth element Gd. The research delves into the impact of Gd addition on phase formation and mechanical properties of high-entropy

alloys, elucidating the enhancement mechanism and deformation behavior of matrix alloys with varying Gd content. This investigation aims to contribute to the advancement of rare earth high-entropy alloy systems with superior performance.

Mechanical properties

The Vickers hardness of $Gd_xCoCrFeNiV_{0.4}$ alloys is illustrated in Fig. 5(a). Gd_0 exhibits only the FCC structure, with a hardness of 177.5 HV. As the volume fraction of the HS phase increases, the hardness value rises from 236.8 HV for the $Gd_{0.1}$ alloy to 410.9 HV for the $Gd_{0.2}$ alloy. The subsequent increase in hardness for the $Gd_{0.3}$ and $Gd_{0.4}$ alloys is attributed to the combined effects of the BCC and HS phases. Specifically, as the volume fractions of both the BCC and HS phases increase, the hardness value increases from 646.4 HV for the $Gd_{0.3}$ alloy to 848.4 HV for the $Gd_{0.4}$ alloy.

The stress-strain curve depicted in Fig. 5(b) illustrates the yield strength, fracture strength, and fracture strain of $Gd_xCoCrFeNiV_{0.4}$ alloys as detailed in Table 4. The yield strength of the alloy initially increases and then decreases. Notably, Gd_0 and $Gd_{0.1}$ alloys exhibit lower yield strength, but show a distinct work hardening stage post-yielding, indicating excellent ductility. These alloys did not fracture even when the strain exceeded 40%. $Gd_{0.3}$ alloy exhibited a yield strength of 1356.17 MPa and a compressive strength of 1620.26 MPa, outperforming the other experimental groups. The fracture strain decreased as the Gd content increased, with $Gd_{0.4}$ exhibiting only 5.68% fracture strain, and lower yield strength and fracture strength compared to $Gd_{0.3}$. The larger atomic radius of Gd contributed to improved solution strengthening effects, along with the formation of the HS phase due to Gd addition. This phase, categorized as a topologically dense heap phase with limited slip systems, further reduced the alloy's plasticity. EBSD analysis revealed that $Gd_{0.4}$ had a larger average grain size than $Gd_{0.3}$, and excessive Gd addition caused pronounced precipitation of V and Cr elements in a BCC structure, leading to the deterioration of mechanical properties in $Gd_{0.4}$ alloy.

The kernel average misorientation (KAM) diagram presented in Fig. 5(c) shows that the FCC phase predominantly governs the deformation process, exhibiting a larger deformation area and higher deformation stress. The HS phase gradually expands the deformation area as the Gd content increases. In the $Gd_{0.3}$ and $Gd_{0.4}$ alloys, the presence of the BCC phase does not contribute to an increase in deformation stress. Overall, the KAM values of the $Gd_{0.3}$ alloy consistently fall within the lower range, indicating fewer defects and excellent overall performance of the specimens.

Gd_0 and $Gd_{0.1}$ exhibited favorable plasticity and did not fracture, which precluded any observation of fracture characteristics. Consequently, a fracture SEM analysis was performed on $Gd_{0.2}$, $Gd_{0.3}$, and $Gd_{0.4}$, with the experimental results illustrated in Fig.

6. Specifically, Fig. 6(d) and (e) present the EDS results for V and Cr elements associated with the fracture of the Gd0.3 alloy, while Fig. 6(f) and (g) show the EDS results for V and Cr elements related to the fracture of the Gd0.4 alloy. The analysis indicated that the fractures of these three alloys exhibited typical brittle characteristics, with river patterns observed in Gd0.2 and the presence of brittle phase particles in Gd0.3 and Gd0.4. These brittle phase particles were identified as the (V, Cr)-rich BCC phase mentioned previously. Furthermore, the stress-strain curve demonstrated a significant reduction in the compressive strength of the Gd0.4 alloy compared to Gd0.3, suggesting that an excessive increase in this brittle phase may be detrimental to enhancing the alloy's strength.