Differential Phase Contrast Microscopy and Atom Probe Tomography of Ferromagnetism in High-Entropy Alloys

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Background incl. aims
High-entropy alloys (HEAs) are emerging as promising materials for future applications due to their high configurational entropy and diverse local atomic environments. Among the four core effects in HEAs, the concept of the 'cocktail effect', resulting from multi-principal alloy design, is particularly intriguing as it can lead to the emergence of new properties. When elements with different structural, kinetic, and ferromagnetic properties are combined in single-phase or nanocomposite HEAs, unexpected interactions and the emergence of new properties or phases can occur, surpassing predictions based on simple mixtures. Certain equiatomic alloys, initially paramagnetic, have been found to exhibit ferromagnetic properties after undergoing deformation processes like rolling or cold-working. This unexpected behavior has been attributed to changes in the local atomic environment and the creation of defects during deformation, resulting in the formation of ferromagnetic clusters. Understanding the mechanisms behind deformation-induced ferromagnetism in these alloys is the aim of this contribution.

Methods
In this study, single phase (CoCrFeMnNi) and nanocomposite HEAs (CoCrFeMnNi and HfNbTaTiZr) were processed using high-pressure torsion (HPT), subjecting the samples to a constant pressure of 9 GPa either as a single disk or stacked disks, with the top anvil rotating at 1 rpm at ambient temperature for up to 15 revolutions. Vibrating sample magnetometry (VSM) confirmed that HPT processing induces the development of ferromagnetic properties. The distribution and orientation of magnetic domains post-deformation were examined in detail using differential phase contrast scanning transmission electron microscopy (DPC STEM), analytical TEM and atom probe tomography (APT) analysis.

Results
Analytical TEM indicates that while the nanocomposite HEA shows significant phase separation due to co-deformation, its saturation magnetization measured by VSM is lower than that of the HPT-processed Cantor alloy. The pronounced phase separation in the nanocomposite HEA leads to lamellar-like alignments of NiCo-rich ferromagnetic and Cr-rich anti-ferromagnetic domains observed by DPC, reducing the overall magnetic moment. Conversely, the absence of such phase separation in the HPT-processed Cantor alloy results in comparable coercivity. APT analyses reveal that this difference arises from the deformation-induced local enrichment of ferromagnetic elements, particularly Ni, showcasing the 'cocktail effect' in HEAs where inter-element interactions lead to unique magnetic behavior.

Conclusions
Our study demonstrates that HPT processing of HEAs induces a transition from paramagnetic to ferromagnetic states at ambient conditions. Deformation-induced ferromag-netism can be explained by the 'cocktail effect' in HEAs, where the formation of ferromagnetic particles is linked to deformation-induced element-selective atomic migration and local enrichment of ferromagnetic elements.

Keywords:

high-entropy alloy; ferromagnetism; nanocomposites; deformation