Feasibility study for the neutron investigation of innovatively manufactured high entropy alloys (HEAs)

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Abstract: High entropy alloys (HEAs) produced via Wire Arc Additive Manufacturing (WAAM) have recently emerged as promising structural materials owing to their design flexibility and potential for controlled chemical composition. Their phase stability and crystallographic texture, two fundamental aspects influencing mechanical behaviour, nevertheless, remain insufficiently characterized at the bulk scale. This feasibility study examines whether neutron diffraction (ND) can effectively probe the microstructure of WAAM-fabricated HEAs. Using an as-printed FeCrCoNiMo+Ta (≤1%) alloy sample, we demonstrate that ND successfully resolves the phase constitution, lattice parameters, peak-broadening features and solidification-induced texture of the material. The results confirm that neutron techniques are well suited for detailed nano(micro)-structural characterization of additively manufactured HEAs and provide a solid foundation for future, more comprehensive investigations.

Keywords: *Neutron diffraction; High Entropy Alloys; Wire arc additive manufacturing, Structure*

1. Introduction

High entropy alloys (HEAs), typically composed of five or more principal elements in near-equimolar ratios, have attracted considerable interest due to their simple crystal structures and exceptional mechanical properties. Cantor-type FCC HEAs such as equimolar CrMnFeCoNi exhibit unusually high toughness and stability, particularly at low temperatures, making them appealing for advanced engineering applications.

Traditional HEA processing routes (powder metallurgy and casting) often involve high energy consumption. As an alternative, Wire Arc Additive Manufacturing (WAAM) offers a cost-effective and versatile approach to fabricating HEAs. WAAM benefits from controllable heat input, industrial accessibility of wire feedstocks, and the ability to generate near-net-shape components. When combined with metal powder-cored wire, WAAM further improves alloying flexibility: metallic powders such as Co, Mn, Cr, and Ni can be incorporated directly into a steel sheath and melted during deposition, forming a high-entropy composition in situ.

Despite these advantages, the microstructural characteristics of WAAM-processed HEAs remain insufficiently understood. Solidification under steep thermal gradients is expected to generate strong crystallographic textures, while the phase stability of Cantor-type alloys under WAAM conditions is still largely unexplored. Bulk-sensitive characterization is therefore essential. ND, owing to its deep penetration and ability to probe large volumes of material, is ideally suited for determining:

- phase constitution
- lattice parameters
- residual-strain indicators
- crystallographic texture.

The present work evaluates whether ND can reliably extract some of these features from WAAM-produced HEAs. As a feasibility assessment, only the as-printed sample is analysed.

2. Materials and methods

The investigated alloy was produced by WAAM using a 2.4-mm-diameter metal powder-cored wire. A low-carbon steel strip was used as the outer sheath, filled with powders of Co, Mn, Cr, and Ni at approximately 20-25% each. During arc melting, the powders and steel sheath combine to form a Cantor-type high-entropy composition. The deposited material was machined into a rectangular block (100×20×25 mm³) suitable for neutron measurements. This geometry enabled the study of bulk texture and phase constitution without sectioning. The as-printed sample WAAM HEA was examined in this feasibility phase.

Neutron diffraction (ND) is a powerful, non-invasive technique for the qualitative and quantitative analysis of phase composition and structural properties of materials at both macroscopic and atomic scales. Compared to X-ray diffraction, neutrons are far more penetrating (by a factor of about 10^3), allowing ND to probe several millimetres into the bulk of a sample rather than only a few micrometres [1]. This capability enables true three-dimensional mapping of internal residual stresses (RS), which arise from manufacturing processes or thermal treatments rather than external loads. Beyond RS determination, ND is widely used to investigate dynamic behaviour, crystallographic texture, phase transformations and internal structural variations without damaging the material. These features have significantly expanded the adoption of neutron-based techniques in recent years, leading to a growing number of industrial applications where bulk analysis is critical for improving production processes, and quality [2-5]. Modern ND experiments also employ high-resolution time-of-flight (TOF) diffractometers with thermal neutron beams and velocity selectors. The TOF-ND approach provides detailed information on structure, phase, texture and sample inhomogeneities, enabling accurate comparison with reference materials and supporting a comprehensive understanding of material’s behaviour.

In the present work, ND was performed using the MEREDIT medium resolution neutron powder diffractometer at NPI Řež, dedicated to studies of atomic and/or magnetic structure of powder polycrystalline samples [6]. Measurements were performed at room temperature for approximately 9 hours and the sample was spinning about its axis during data collection. The adopted slit size was 10×10 mm2. A monochromator was used, having the following setup: mosaic Cu, Reflection (220) - symmetric, wavelength λ = 1.460 Å. Data were collected over a 2θ range from 4° to 144° with a step of 0.08°. Raw data were processed to extract peak positions, full width at half maximum (FWHM) values and crystallographic texture.

3. Results and Discussion

The diffraction pattern of Figure 1, corresponding to the as-printed WAAM high entropy alloy sample, represents the raw ND intensity as a function of the diffraction angle 2θ over a wide angular range (approximately, 4-143°).

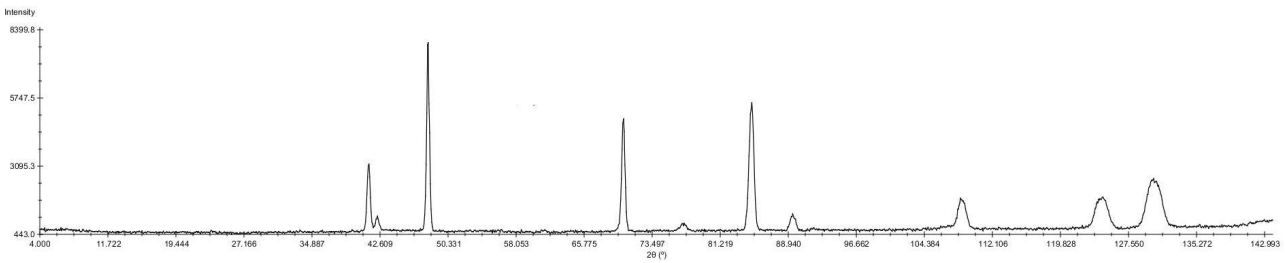


Figure 1. ND pattern of the as-printed WAAM high entropy alloy sample.

The background level appears low and relatively flat, indicating good data quality and limited incoherent scattering. A series of sharp and intense Bragg peaks, superimposed on this background, demonstrates that the considered material is well crystallized despite being produced by WAAM.
The most prominent reflections are observed at approximately 2θ ≈ 42-43°, 48-50°, 70-71° and 84-85°, corresponding to the (111), (200), (220) and (311) planes of a face-centred cubic (FCC) lattice, respectively. The dominance of these peaks confirms that the as-printed alloy is notably single-phase FCC, consistent with Cantor-type high entropy alloys. A very small amount of high-angle residual intensity suggests, at most, the presence of trace BCC-like features, but no additional strong or systematic peaks incompatible with the FCC structure are detected.
The (200) reflection is particularly intense, indicating a pronounced preferred crystallographic orientation. This intensity enhancement reflects a strong texture induced by directional solidification during the WAAM process, where steep thermal gradients promote epitaxial grain growth along specific crystallographic directions.
From the position of the (200) peak at approximately 48.08°, the FCC lattice parameter was estimated as a ≈ 3.59 Å, in good agreement with values reported for Cantor-type alloys. Although the diffraction peaks are generally narrow, they exhibit exhibit measurable broadening, especially at higher angles. The full width at half maximum (FWHM) values range from approximately 0.36° to 0.63°, depending on the reflection. Such broadening is typical of rapidly solidified additively manufactured materials and can be attributed to a combination of residual stresses imposed by steep WAAM thermal gradients, high dislocation densities and the presence of sub-grain or cellular structures formed during deposition.
The ability to detect and quantify this peak broadening in a relatively large bulk specimen demonstrates that ND can access microstructural indicators essential for evaluating WAAM process quality, including RS and strain state, defect density and solidification-induced substructures.
The crystallographic texture of the as-printed WAAM high entropy alloy was investigated by neutron diffraction through the reconstruction of pole figures (PFs) and inverse pole figures (IPFs) for the FCC phase. The texture analysis reveals a pronounced preferred orientation induced by the additive manufacturing process.

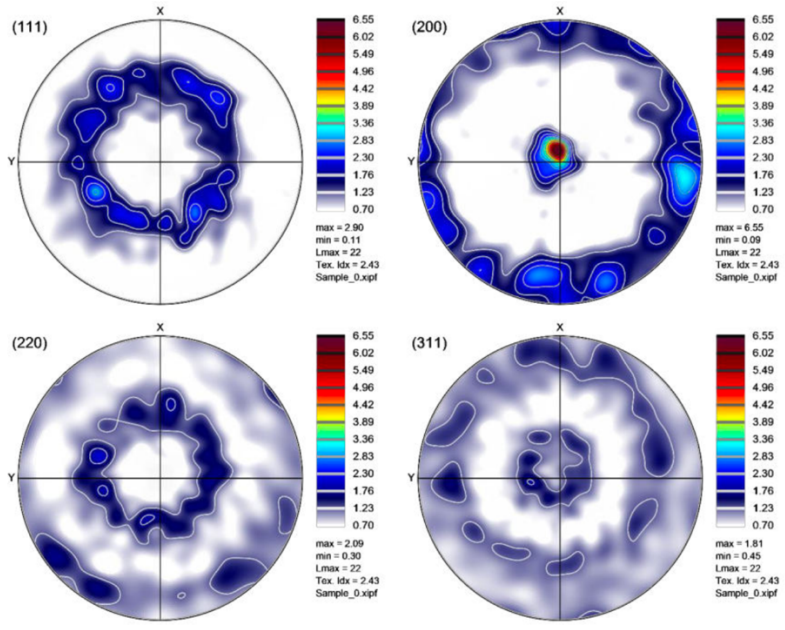


Figure 2. Pole figures

The FCC (200) pole figure (fig.2) exhibits a strong intensity maximum at the centre of the stereographic projection, whereas the (111), (220), and (311) pole figures show characteristic ring-like intensity distributions at specific tilt angles. This combination of features is typical of a 〈001〉 fibre texture, in which the crystallographic 〈001〉 direction is preferentially aligned with the build

direction of the component. Such a texture is a well-known signature of directional solidification in FCC materials processed under steep thermal gradients.

The observed distribution is consistent with a well-developed ⟨001⟩ fibre texture, typical for FCC materials solidified under directional thermal gradients. The relatively continuous intensity along the rings suggests a certain spread of orientations around the fibre axis, rather than a sharp single-crystal-like alignment. The maximum texture intensity reaches approximately 6.5 m.r.d. in the (200) pole figure, confirming a strong but not fully ideal fibre component.

Such texture development is characteristic of WAAM processing, where epitaxial grain growth along the direction of heat extraction promotes preferential selection of grains with ⟨001⟩ orientation parallel to the build direction. The pole figure analysis therefore provides direct experimental evidence supporting the texture-induced anisotropy inferred from the diffraction peak intensity distribution.

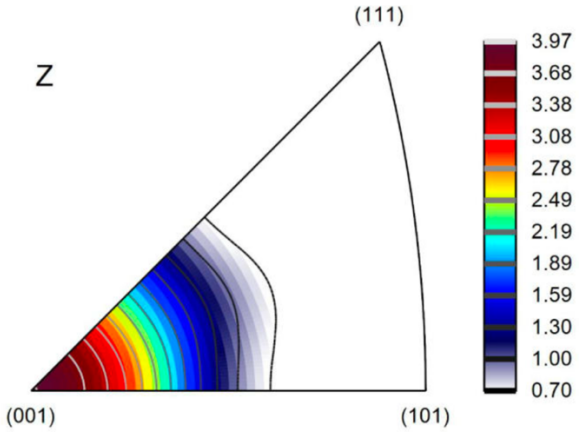


Figure 3. Inverse pole figure

The inverse pole figures (fig.3) further corroborate this interpretation. The IPF corresponding to the build direction (Z) displays a pronounced intensity concentration near the (001) corner of the FCC fundamental triangle, with a maximum texture strength of approximately 4 multiples of a random distribution. In contrast, the IPFs associated with the transverse directions (X and Y) are significantly more diffuse, showing broader orientation spreads extending from the (001)-(101) edge toward the interior of the triangle. This indicates that crystallographic alignment is strongly constrained along the build direction, while the in-plane orientations are comparatively weakly defined.

The presence of a strong 〈001〉 fibre texture is fully consistent with the thermal conditions inherent to the WAAM process. During deposition, the steep and directional heat flow promotes epitaxial grain growth from previously solidified material, favouring FCC grains whose 〈001〉 direction aligns with the direction of maximum heat extraction. The unusually high intensity of the (200) reflection observed in the ND pattern (Fig. 1) directly reflects this texture component and provides independent confirmation of the orientation analysis.

Overall, the texture results demonstrate that ND is capable of reliably quantifying solidification-induced crystallographic texture in bulk WAAM-fabricated high entropy alloys. The identification of a strong 〈001〉 fibre texture in the as-printed condition highlights the intrinsic anisotropy introduced by the manufacturing process and underscores the importance of texture-sensitive bulk characterization carried out in order to understand the structural and mechanical behaviour of additively manufactured HEAs.

4. Conclusion

This feasibility study demonstrates that ND is a robust and insightful tool for analysing the microstructure of high entropy alloys produced by WAAM. Using the as-printed specimen, we establish that:

- the alloy is predominantly single-phase FCC with a stable lattice parameter
- peak shapes reveal residual-strain and solidification microstructures
- a distinct〈001〉fibre texture is clearly quantified along the build direction.

These findings validate ND as a suitable methodology for studying phase stability and crystallographic texture in WAAM-fabricated HEAs and provide a strong basis for future, more extensive investigations. The basis for future quantitative work is founded by current research.

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