

Reviewer 2

We sincerely thank the Reviewer for the positive evaluation of our manuscript and for the constructive suggestions. All comments have been carefully considered, and the manuscript has been revised accordingly. Our detailed responses are provided below.

The present manuscript describes a combined approach using CALPHAD thermodynamic modelling and experimental characterization to investigate the effect of boron content on the formation of boride phases in the Fe-Ni-Cr-Cu-Si-B-C alloy system. The topic is timely and relevant to the development of wear-resistant coatings and multicomponent Fe-based alloys. The authors identify an optimal boron content (~4 wt.%) and an optimal processing temperature (638 °C) for achieving a stable multiphase microstructure with a significant fraction of the Fe₂B phase.

However, the optimal boron content is determined solely by CALPHAD calculations, whereas the experimental characterization is performed for only a single alloy composition. This limitation should be clearly stated in the Abstract. The current wording is somewhat misleading, as it may give the impression that several compositions were experimentally investigated.

Response: We thank the Reviewer for this valuable comment. We agree that the original wording could give the impression that multiple alloy compositions were experimentally investigated. The Abstract has been revised to clarify that the optimization of boron content was performed using CALPHAD calculations and response surface analysis, whereas the experimental characterization was carried out only for the selected optimized composition (4 wt.% B). This limitation has also been explicitly discussed in the newly added "Limitations of the Present Study" section.

The manuscript is not consistent in its terminology. The investigated material is referred to both as a high-entropy alloy (HEA) and as an Fe-based alloy, although the alloy contains a higher Cu content than Fe. The terminology should be unified throughout the manuscript, or the classification should be properly justified.

Response: We thank the Reviewer for this valuable comment. We agree that the investigated material should not be classified as a high-entropy alloy according to the generally accepted criteria. Accordingly, the manuscript has been revised throughout, and the terminology has been unified by consistently referring to the investigated material as a Fe-based multicomponent alloy. References to high-entropy alloys have been retained only where they provide general scientific background.

The description of the experimental methods is inconsistent. X-ray diffraction (XRD) is not mentioned in the Abstract despite being an important characterization technique.

Response: We thank the Reviewer for pointing this out. The Abstract has been revised to explicitly include X-ray diffraction (XRD) among the experimental characterization techniques employed in this study.

Furthermore, the Experimental section states that wavelength-dispersive spectroscopy (WDS) was employed, whereas the Results section refers to EDS data. This inconsistency should be resolved. In particular, the determination of boron and carbon by EDS is problematic due to the limited accuracy of this technique for light elements.

Response: We thank the Reviewer for identifying this inconsistency. The manuscript has been carefully revised to ensure consistent terminology throughout. Since the experimental characterization was performed using SEM-WDS, all incorrect references to EDS have been removed from the manuscript.

The Experimental section should specify the chemical form in which boron was introduced into the powder mixture.

Response: We thank the Reviewer for this valuable comment. The Experimental section has been revised to clarify the chemical form of boron used during alloy preparation. Boron was introduced into the powder mixture as a nickel–boron master alloy (NiB_{15}), while the remaining alloying elements were added as elemental powders (Fe, Ni, Cr, Cu, Si, and C).

In addition, were the contributions of carbon and silicon originating from the liquid glass binder included in the nominal alloy composition and thermodynamic calculations?

Response: Liquid glass (5 wt.% aqueous solution) was used exclusively as a temporary inorganic binder during the agglomeration of the mechanically activated powder mixture. The solvent was removed during drying (80–100 °C), followed by sintering of the agglomerated granules at 600 °C and subsequent milling prior to spraying. Consequently, the nominal alloy composition and the CALPHAD calculations were based on the metallic powder mixture only. The contribution of the binder to the overall alloy composition was not considered, as it served solely a technological function during powder preparation. This clarification has been added to the Experimental section of the revised manuscript.

Since oxygen was used as the spraying gas, was the presence of oxide phases investigated or detected in the deposited coatings?

Response: We thank the Reviewer for this valuable comment. We agree that oxidation may occur during gas-flame spraying because oxygen is used as the oxidizing gas. However, the present study did not specifically investigate oxide formation. No distinct oxide phases were identified by XRD, and the SEM-EDS analysis did not provide conclusive evidence for the presence of oxide phases. Therefore, oxide formation cannot be completely excluded, particularly at levels below the detection limits of the employed techniques. This limitation has been acknowledged in the revised manuscript and identified as a subject for future investigation using dedicated phase- and chemistry-sensitive techniques.

In Figure 2, it should be more clearly emphasized that the presented phase fractions are CALPHAD predictions rather than experimentally verified results.

Response: The figure and the corresponding caption have been revised to clearly indicate that all phase fractions shown in Figure 2 represent CALPHAD equilibrium predictions rather than experimentally determined phase fractions.

In addition, presenting the results as four separate plots instead of two combined figures would considerably improve readability.

Response: We appreciate this suggestion. However, to preserve the logical comparison between the investigated variables and to avoid substantially increasing the figure size, we retained the original layout. Nevertheless, the figure resolution, labels, and caption have been improved to enhance readability.

The prediction of three coexisting liquid phases is unusual and deserves a more detailed discussion. In this respect, experimental thermal analysis (DSC or DTA) would provide valuable validation of the calculated phase transformations and should be considered or at least discussed as a limitation of the present study.

Response: We agree that the prediction of multiple liquid phases is an interesting thermodynamic feature. A short discussion has been added to the manuscript emphasizing that these phase fields represent equilibrium CALPHAD predictions. We also agree that DSC/DTA experiments would provide valuable experimental verification of the predicted phase transformations. Since such experiments were beyond the scope of the present study, this aspect has been acknowledged in the newly added "Limitations of the Present Study" section and identified as an important direction for future work.

The scale bars in the SEM micrographs should be enlarged or otherwise made more visible.

Response: The scale bars have been enlarged to improve readability.

The elemental maps appear blurred. Presenting them in a monochromatic colour scale may improve their clarity and facilitate interpretation.

Response: The elemental maps are now presented using a monochromatic color scale as suggested by the Reviewer.

Figure 8 requires a more detailed description. What do the individual data points and their error bars represent? Furthermore, does connecting the points with lines have any physical meaning, or would another type of graphical representation be more appropriate?

Response: We thank the Reviewer for this valuable comment. Figure 8 has been revised to improve its clarity. The individual data points represent independent Vickers hardness measurements performed at different locations within the coating, diffusion zone, and steel substrate. The error bars correspond to the standard deviation of the measurements in each region, while the values labeled "mean" represent the average hardness. Since the indentation positions are independent measurements rather than a continuous variable, the connecting lines have been removed to avoid implying a physical trend. The figure caption and the corresponding discussion in the manuscript have been revised accordingly.

Finally, the presence and morphology of fine precipitates cannot be conclusively demonstrated by SEM alone. If the authors wish to discuss nanoscale precipitates, additional characterization by transmission electron microscopy (TEM) or another high-resolution technique (e.g., small-angle scattering) would be necessary.

Response: We fully agree with the Reviewer. Accordingly, the discussion has been revised to avoid definitive statements regarding nanoscale precipitates. Since TEM or other high-resolution characterization techniques were not performed, the interpretation has been limited to the microstructural features directly supported by SEM observations. This limitation has also been acknowledged in the revised manuscript.

The claimed agreement between CALPHAD predictions and experimental observations should be moderated. The presented SEM, EDS/WDS, and XRD results provide only qualitative evidence of the predicted phases. Without quantitative phase analysis (e.g., Rietveld refinement of the XRD data), it is not possible to conclude that the thermodynamic predictions have been fully validated.

Response: We thank the Reviewer for this valuable comment. We fully agree and have revised the manuscript accordingly. Throughout the revised manuscript, the agreement between the CALPHAD calculations and the experimental observations is consistently described as

qualitative rather than quantitative. The terminology has been revised to avoid overly strong claims regarding experimental validation of the thermodynamic predictions.

