Data-driven design of hydrogen solubilities in metallic alloys

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Establishing hydrogen economy to reduce carbon emission makes understanding mechanisms for hydrogen embrittlement and hydrogen storage in metallic alloys of great interest. One key goal in this regard is to control the amount of hydrogen that can be incorporated into a metallic crystral structure, as determined by the hydrogen (H) solution enthalpy.

In a multistage analysis, first the solution enthalpy of H in the 3d transition metals has been systematically studied by density functional theory (DFT) [1]. A universal dependence on chemical and geometric parameters has been observed [2]. The study has been extended to other interstitial elements and transition metals. Applying a principle component analysis (PCA) to the large dataset, we found that interstitial solution enthalpies can be described by up to three common factors (Figure 1). Similar to the interstitial elements C, N, and O, the solubility of H is determined by nearest-neighbor atomic distances and the electronic density of states of the transition metals. The outcome was used to derive a master curve for the solubility of interstitials in metals.

In a second step, we systematically analyze with DFT the impact of M = Cr, Mn, Fe and their mixtures on the H solubility for two carbide phases, M_3C and $M_{23}C_6$, which are important precipitates in steels [3]. Our results show that the addition of these alloying elements yields strong nonmonotonic chemical trends for the H solubility. We identify magneto-volume effects as the origin for this behavior, which depend on the considered system, the sites occupied by H, and short- vs long-range interactions between H and the alloying elements. We further show that the H solubility is directly correlated with the occupation of its nearest-neighbor shells by Cr and Mn.

Based on these insights, DFT data from H containing binary-metal carbides are used to design a ridge regression-based model that predicts the solubility of H in the ternary-metal carbides (Figure 2). In a final step the reasoning abilities of large-language model (LLMs) are explored in this context. The LLM is asked to use experimental data, and explicate and implicit knowledge about the carbides to predict the H solution enthalpy for composition ranges that were not part of the training set. The work affirms that performing the reasoning of the LLM in terms of programming code leads to well performing and transparent models for the prediction task [4].

References

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Figures

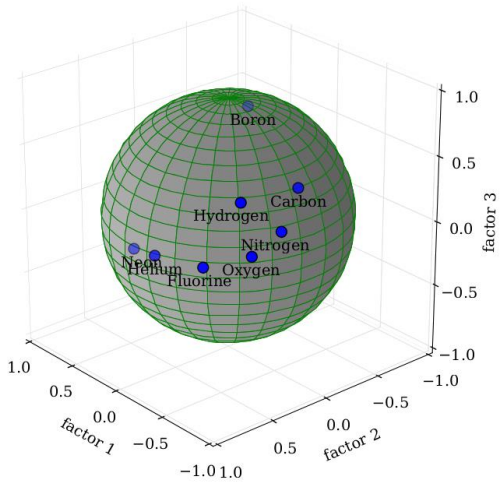

Figure 1. Results of the principle component analysis (after the reduction to three factors) for the solubility of interstitial elements in 3d transition metals [1].

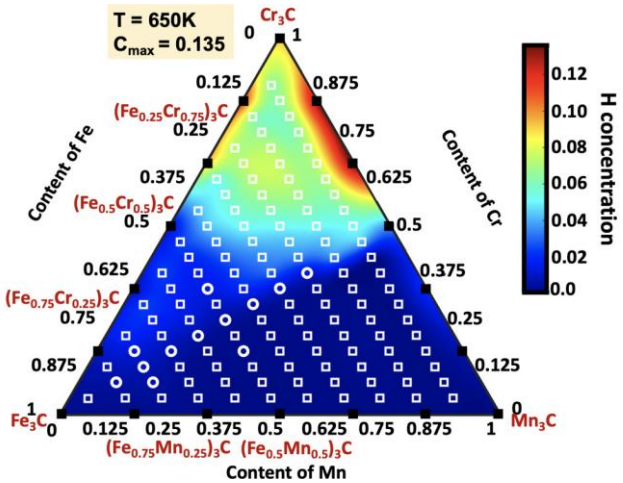

Figure 2. Heat maps of H concentration in (Fe,Cr,Mn)3C carbides at 650 K. The white nonfilled squares represent H concentration values in ternary metal carbides predicted by the machine learning model.