Tracer diffusion in the Ni–CoCrFeMn system: Transition from a dilute solid solution to a high entropy alloy

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Goal:

- study the influence of lattice structure and microstructure on the thermomechanical properties
  - two families of HEA, fcc and bcc including their coexistence, namely the $Co_xCr_xFe_xMn_xNi_{100-4x}$ and the $Al_{100-4x}Co_xCr_xFe_xNi_x$
- Structural discontinuities? Changes upon compositional variations? Underlying physical principles?

Approach:

- variation of the alloying content by "continuously" varying the amount of components until an equiatomic composition
- Wu et al.[1]: successively increased number of alloying constituents

[1] Z. Wu et al. In: *Intermetallics* (2014).

Appearance of the HEA effect

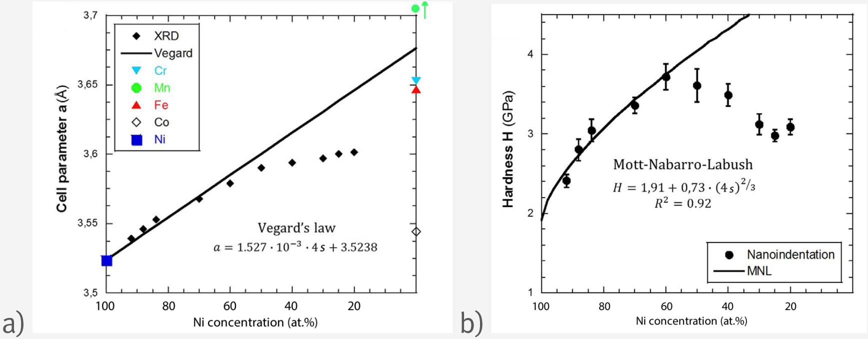


Figure: Lattice parameter and hardness behaviour of $(CoCrFeMn)_{100-x}Ni_x$.[2]

[2] Mathilde Laurent-Brocq et al. In: *Materials Science and Engineering: A* (2017).

Investigated system

- $Co_xCr_xFe_xMn_xNi_{100-4x}$
- two dilute solid solutions:
  - $Co_2Cr_2Fe_2Mn_2Ni_{92}$
  - $Co_{10}Cr_{10}Fe_{10}Mn_{10}Ni_{60}$
- one high entropy alloy:
  - $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$
- Diffusion measurements via radiotracer method

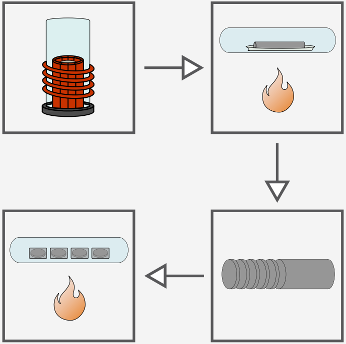
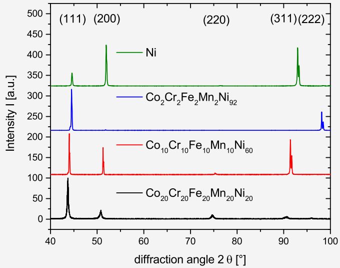
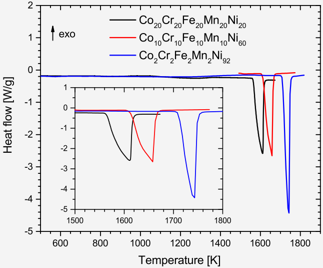


Figure: Steps of sample processing.

Sample properties



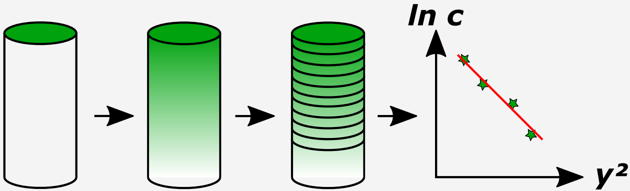
a)



b)

Figure: a) Normalized X-ray diffraction patterns and of $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$, $Co_{10}Cr_{10}Fe_{10}Mn_{10}Ni_{60}$, $Co_2Cr_2Fe_2Mn_2Ni_{92}$ and pure Ni. b) DSC curves of $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$, $Co_{10}Cr_{10}Fe_{10}Mn_{10}Ni_{60}$ and $Co_2Cr_2Fe_2Mn_2Ni_{92}$ alloys.

Diffusion measurements - Radiotracer method



- instantaneous source solution of the diffusion problem[3]

$$c(y,t) = \frac{M}{\sqrt{\pi D_{\mathrm{V}} t}} \exp\left(-\frac{y^2}{4 D_{\mathrm{V}} t}\right) \tag{1}$$

- Gaussian solution of the diffusion problem:

$$D_{\mathrm{V}} = \frac{1}{4t}\left(-\frac{\partial \ln c(y,t)}{\partial y^2}\right)^{-1} \tag{2}$$

[3] A. Paul et al. Springer, 2014.

Summary of diffusion measurements

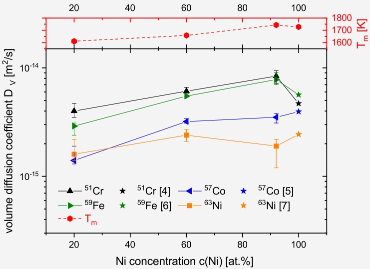


Figure: Measured volume diffusion coefficients D_V at 1373 K in the $(CoCrFeMn)_{100-x}Ni_x$ alloys and Ni.[4][5][6][7]

- Diffusion of Co, Cr and Fe in $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$ is slower than in $Co_2Cr_2Fe_2Mn_2Ni_{92}$
- Differences are comparable with those between pure Ni and $Co_2Cr_2Fe_2Mn_2Ni_{92}$
- Changes of diffusion coefficients are within a factor of three (Co) or even marginal (Ni).

[4] K. Monma, H. Suto, and H. Oikawa. In: *J. Jpn Inst. Metals* (1964)
[5] A. Vladimirov et al. In: *Fizika Metallov i Metallovedenie* (1978)
[6] H. Bakker, J. Backus, and F. Waals. In: *physica status solidi (b)* (1971)
[7] M. Bronfin, G. Bulatov, and I. Drugova. In: *Fiz. met. metalloved.* (1975)

Comparison for $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$

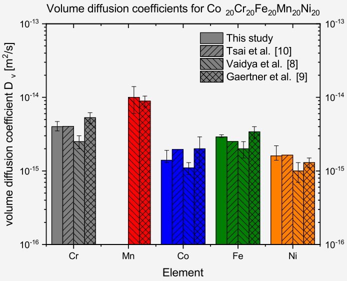


Figure: Comparison of volume diffusion D_V measured at 1373 K in the $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$ alloy.

- Radiotracer diffusion on polycrystalline alloys (this study, Vaidya et al. [8])
- Radiotracer diffusion on a single crystalline alloy (Gaertner et al. [9])
- Diffusion couple consisting of polycrystalline alloys (Tsai et al. [10])

[8] M. Vaidya et al. In: *Acta Materialia* (2018)
[9] D. Gaertner et al. In: *Journal of Materials Research* (2018)
[10] K.-Y. Tsai, M.-H. Tsai, and J.-W. Yeh. In: *Acta Materialia* (2013)

Comparison on homologous temperature scale T_m / T

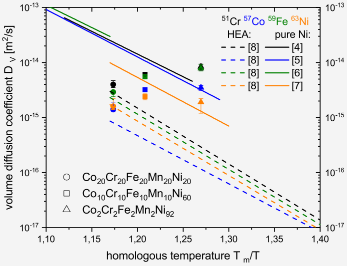


Figure: Volume diffusion coefficients of Cr, Co, Fe and Ni on a homologous temperature scale T_m / T. [4][5][6][7][8]

- Diffusion rates in $Co_{20}Cr_{20}Fe_{20}Mn_{20}Ni_{20}$ are by about one order of magnitude slower than in $Co_2Cr_2Fe_2Mn_2Ni_{92}$
- Diffusion coefficients for $Co_2Cr_2Fe_2Mn_2Ni_{92}$ are within a factor of two of those measured for pure Ni

[4] K. Monma, H. Suto, and H. Oikawa. In: *J. Jpn Inst. Metals* (1964)
[5] A. Vladimirov et al. In: *Fizika Metallov i Metallovedenie* (1978)
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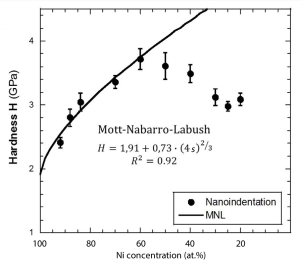
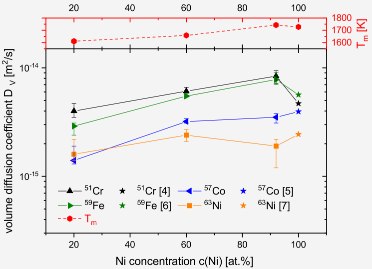
[8] M. Vaidya et al. In: *Acta Materialia* (2018)

Conclusion

- all measured tracer diffusion coefficients in the range of 10^{-15} to 10^{-14} m^2s^{-1}
  - Cr fastest element, followed by Fe, Co and Ni (slowest) at the temperature under investigation
- Overall small changes of diffusion coefficients at 1373 K
  - no indication for sluggish diffusion behaviour
- Comparison at homologous temperatures indicates a slow down of diffusion rates coming from an dilute solid solution and going into high entropy regimes

Comparison mechanical and kinetic properties





- Deviations in mechanical properties[2] are not distinctly shown in volume diffusion measurements[4][5][6][7]

[2] Mathilde Laurent-Brocq et al. In: *Materials Science and Engineering: A* (2017)

[4] K. Monma, H. Suto, and H. Oikawa. In: *J. Jpn Inst. Metals* (1964)

[5] A. Vladimirov et al. In: *Fizika Metallov i Metallovedenie* (1978)

[6] H. Bakker, J. Backus, and F. Waals. In: *physica status solidi (b)* (1971)

[7] M. Bronfin, G. Bulatov, and I. Drugova. In: *Fiz. met. metalloved.* (1975)

Acknowledgment

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DFG Deutsche Forschungsgemeinschaft