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Tracer diffusion in single crystalline CoCrFeNi and CoCrFeMnNi high entropy alloys

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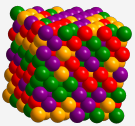
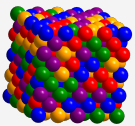
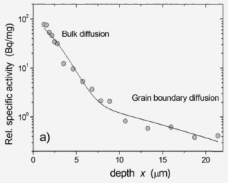
CERN

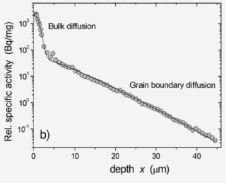
ISOLDE Solid State Physics

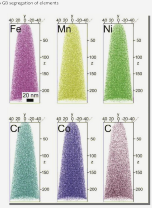
Motivation



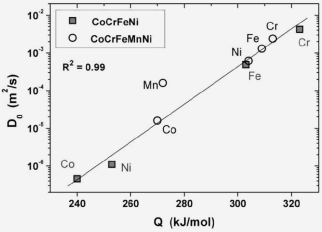








- High-entropy alloys have four or more components in equiatomic proportion
- A “core effect” based on interdiffusion measurements [1]: “sluggish diffusion”
- Shape of first measured penetration profiles are not fully understood yet [2, 3, 4]
- **First tracer diffusion experiments in singlecrystalline FCC HEAs**

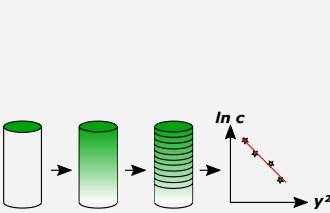


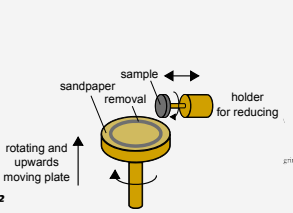
Daniel Gaertner

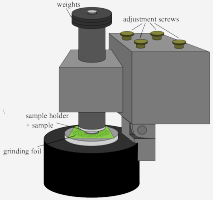
[1] J.W. Yeh et al. Adv. Eng. Mater **6**, (2004)
[2] M. Vaidya et al. JALCOM **688**, (2016)
[3] M. Vaidya et al. Scientific Reports **7**, (2017)
[4] M. Vaidya et al. Acta Mater **146**, (2018)

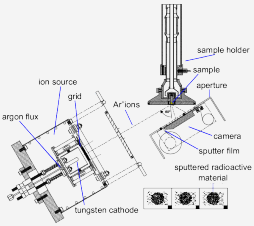
Radiotracer technique

- Homogenized at 1373 K (1100 °C) for 3 days
- Diffusion-annealed at 1373 K (1100 °C) for different times - sectioned by mechanical grinding
- Diffusion annealed at 1063 K (790 °C) - sectioned by ion-beam sputtering





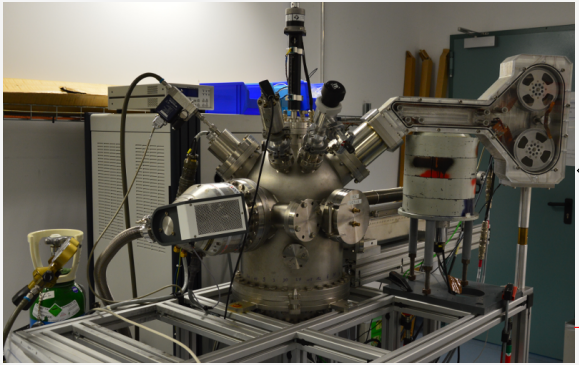


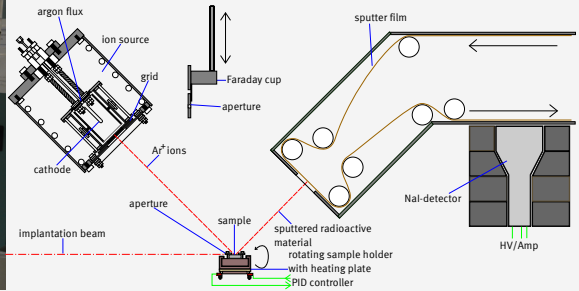


- Gaussian-solution: $D_v = -\frac{1}{4t}\left(\frac{\partial \ln \bar{c}}{\partial y^2}\right)^{-1}$

Radiotracer technique - ISOLDE/CERN

- Cu-diffusion: annealed at 973 K (700 °C) for 12 hours, at 1073 K (800 °C) for 2 hours and at 1173 K (900 °C) for 20 minutes





EDX analysis & orientation

Element	CoCrFeNi at %	CoCrFeMnNi at %
Co	25.1	19.9
Cr	24.9	19.4
Fe	25.2	19.6
Mn	–	20.2
Ni	24.8	20.9

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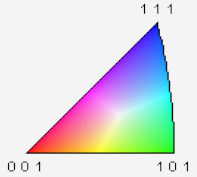
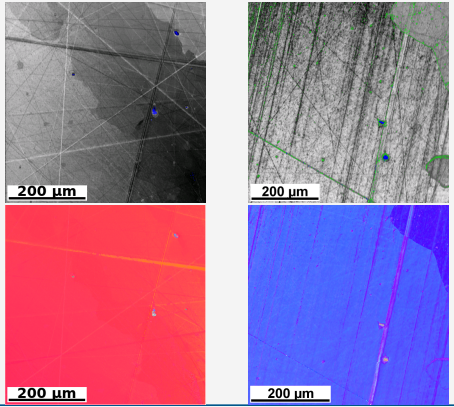
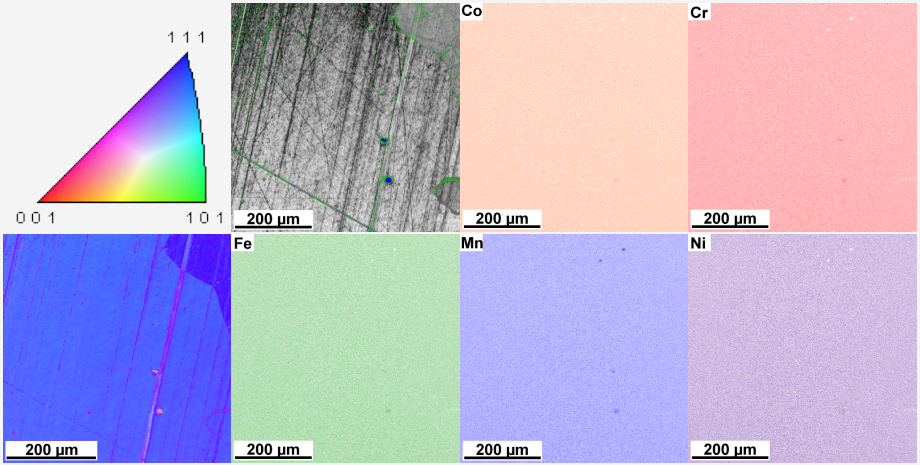


CoCrFeMnNi

⟨001⟩ ⟨111⟩

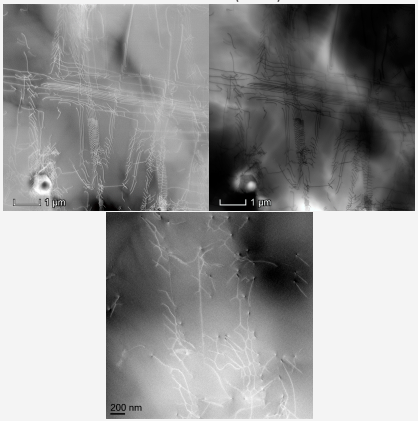


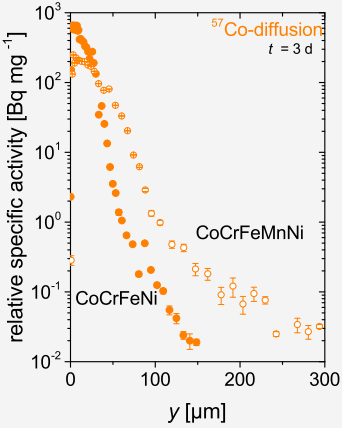
CoCrFeMnNi ⟨111⟩ microstructure



Preliminary TEM-analysis

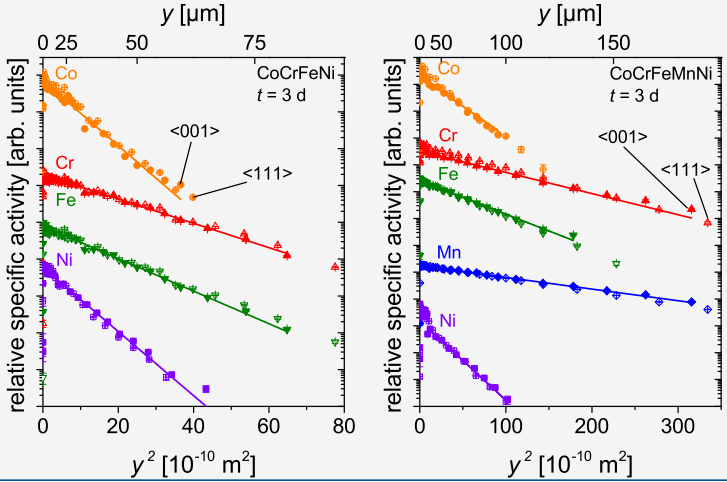
CoCrFeNi ⟨111⟩





- network of dislocation bonds
- contribution of dislocation diffusion
- analysis of dislocation contribution requires more specific TEM-analysis (ongoing work)

Penetration profiles - isotropy and time-dependency



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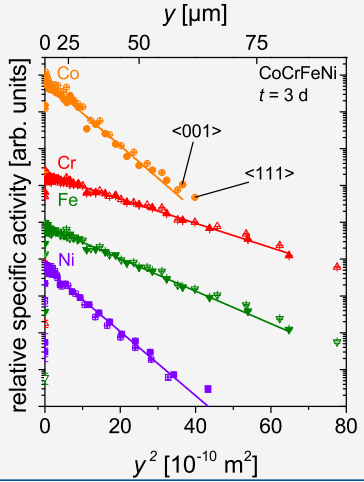

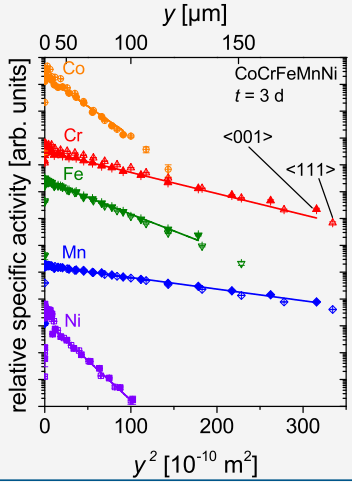

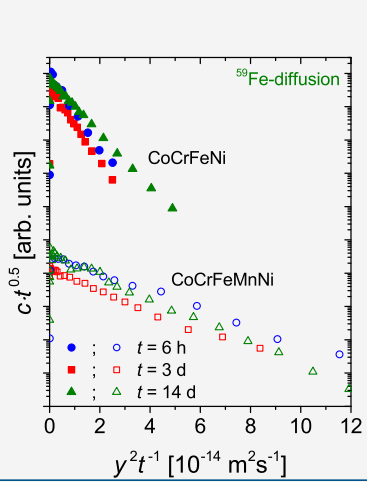

CoCrFeNi

Element	$T = 1373$ K D_V 10^{-15} m^2 s^{-1}	$T = 1063$ K D_V 10^{-19} m^2 s^{-1}
Co	0.5	1.5
Cr	1.8	0.3
Fe	1.1	4.0
Mn	3.0	0.1
Ni	0.4	–

CoCrFeNi

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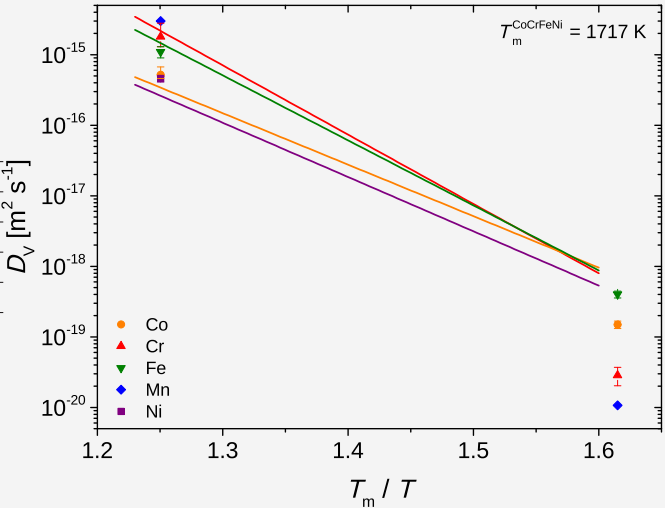
- symbols: singlecrystalline data (present work)
- lines: polycrystalline data [2, 3, 4]



[2] M. Vaidya et al. JALCOM **688**, (2016)
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CoCrFeMnNi

Element	$T = 1373$ K D_V 10^{-15} m^2 s^{-1}	$T = 1063$ K D_V 10^{-19} m^2 s^{-1}
Co	2.0	2.2
Cr	5.3	11.6
Fe	3.4	6.9
Mn	8.9	46.6
Ni	1.3	0.6

CoCrFeMnNi

Element	$T = 1373$ K D_V 10^{-15} m^2 s^{-1}	$T = 1063$ K D_V 10^{-19} m^2 s^{-1}
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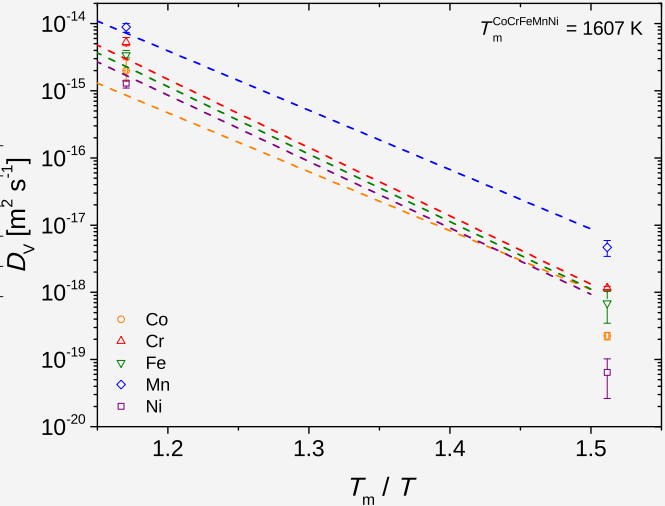

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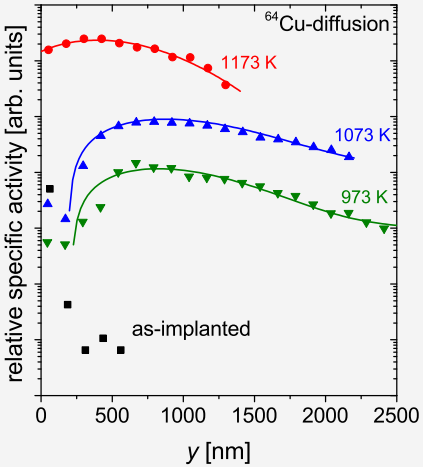
Cu-diffusion in CoCrFeNi



Strohm solution*:

$$\tilde{c}(y,t) = \frac{M_0/2}{\sqrt{1+(2D_V t/\sigma^2)}} \cdot \left[\operatorname{erfc}\left(\frac{-(y_0/2\sigma^2) - (y/4D_V)}{\sqrt{(1/2\sigma^2)+(1/4D_V)}} \right) \cdot \exp\left(\frac{-(y-y_0)^2}{2\sigma^2 + 4D_V t} \right) \right.$$

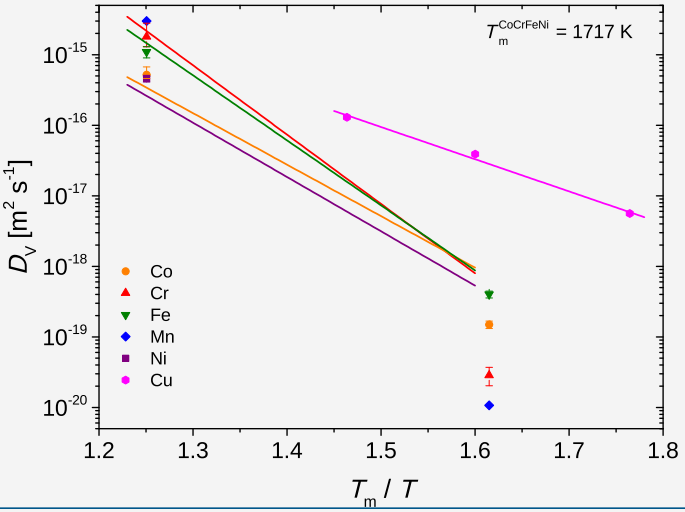
$$\left. + k\operatorname{erfc}\left(\frac{-(y_0/2\sigma^2) + (y/4D_V)}{\sqrt{(1/2\sigma^2)+(1/4D_V)}} \right) \cdot \exp\left(\frac{-(y+y_0)^2}{2\sigma^2 + 4D_V t} \right) \right]$$

y_0: implantation depth; σ^2: width of implanted tracer distribution;
k: surface conditions (≈ -1, surface acts as perfect sink)

T	t	D_V
K	s	$m^2\ s^{-1}$
1173	1200	$1.3 \cdot 10^{-16}$
1073	7200	$3.9 \cdot 10^{-17}$
973	43200	$5.6 \cdot 10^{-18}$

[5] A. Strohm et al. Z. Metallk. **93**, (2002)

Arrhenius plot



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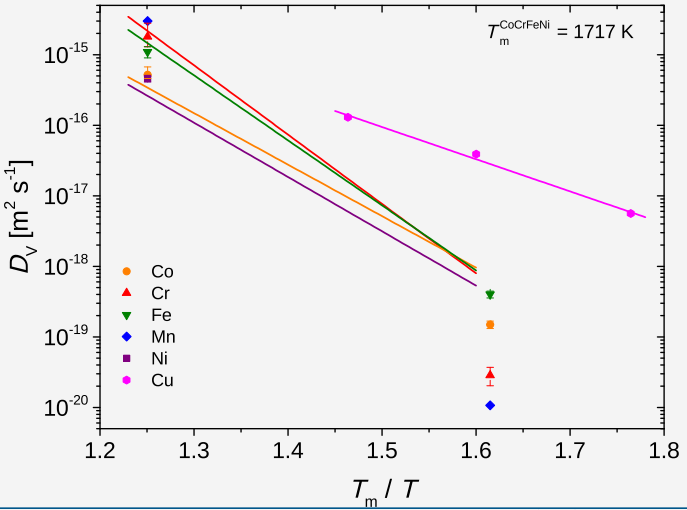
Arrhenius plot



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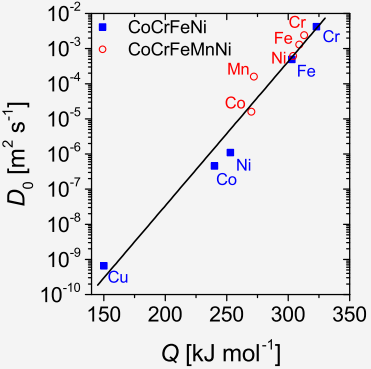

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Summary

- Bulk diffusion results in single crystals are in good agreement with bulk diffusion results in polycrystalline FCC HEAs (at 1373 K)
- Polycrystalline data overestimate the bulk diffusion coefficient at 1063 K
- Diffusion is time-independent and isotropic
- Mn is the fastest element, Ni the slowest element in both systems at 1373 K and also at 1063 K in the quinary alloy
- At 1063 K, Mn and Cr are the slowest elements in the quaternary alloy and Fe the fastest
- Tracer diffusion is not retarded (”sluggish”) with addition of a further alloying element in equiatomic proportion
- Cu is fastest element at 1173 K and below (low activation enthalpy)

Outlook

- Arrhenius-dependence of self-diffusion in CoCrFeNi and CoCrFeMnNi single crystals (measurements planned at 1253 K, 1153 K, 993 K and 923 K)
- Mn-diffusion in CoCrFeNi single crystals
- Grain-boundary diffusion in CoCrFeNi and CoCrFeMnNi bicrystals (ongoing work)
- TEM analysis of the dislocation network in order to analyze the dislocation contribution in the penetration profiles (ongoing work)

Acknowledgements

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DFG Deutsche Forschungsgemeinschaft

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