Supplementary Material

Combining thermodynamics with tensor completion techniques to enable multicomponent microstructure prediction

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This document contains an overview of the results obtained from the 1D and 2D simulations for the alloy compositions given in the table.

Alloys composition			
Alloy	x_1	x_2	x_3
1	0.44	0.10	0.22
2	0.23	0.20	0.40
3	0.30	0.25	0.30
4	0.35	0.30	0.25
5	0.24	0.15	0.47
6	0.46	0.30	0.15

The following information is presented:

- The system definition of each alloy, with the composition, temperature and system size used for the equilibrium calculations.
- A ternary section of the phase-diagram, with the varying compositions indicated along the axes and the present phases in the legend. In all cases, the molar fraction of copper is taken as constant.
- The equilibrium conditions of the two coexisting liquid phases, showing their equilibrium mole fraction and the equilibrium molar fraction of each component.
- The molar fraction fields of Ag, Cu and Ni as obtained from the simulations after 10^5 time steps. In the first row, the molar fraction fields obtained from the simulations using the direct implementation of the CALPHAD expression (CE) are presented; the following rows show the molar fraction fields obtained from simulations using TTMs with rank value $R = 3, \ldots, 10$, as indicated in the text box at the top of the images.
- The probability density function (PDF) of the molar fraction fields after 10^5 time steps. The PDFs are also displayed for the direct implementation with the CALPHAD expression in the first row and for all rank values, in the following rows.
- The volume fraction of one of the liquid phases as a function of the simulation time step.
- The results from the 1D simulation are discussed at the end of the document, for all alloy compositions together and using simulations where the thermodynamic quantities are provided via a programming interface with TC-Toolbox as a reference.

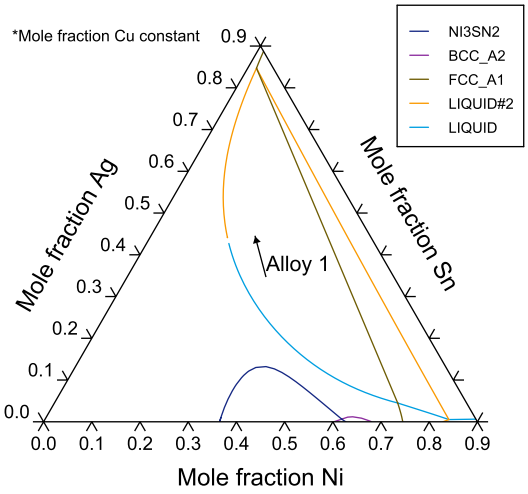
All results presented here are obtained with the phase-field model and the parameters given in the main text. The thermodynamic equilibrium conditions and phase-diagrams are calculated using Thermo-Calc 2017b and the COST531 database. The PDFs are obtained according to the *Langer-Bar-On-Miller* (LBM) method.

Alloy 1

System definition

Component	Mole Fraction	Mass fraction
Ag	0.44	0.498
Cu	0.10	0.067
Ni	0.22	0.135
Sn	0.24	0.229

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

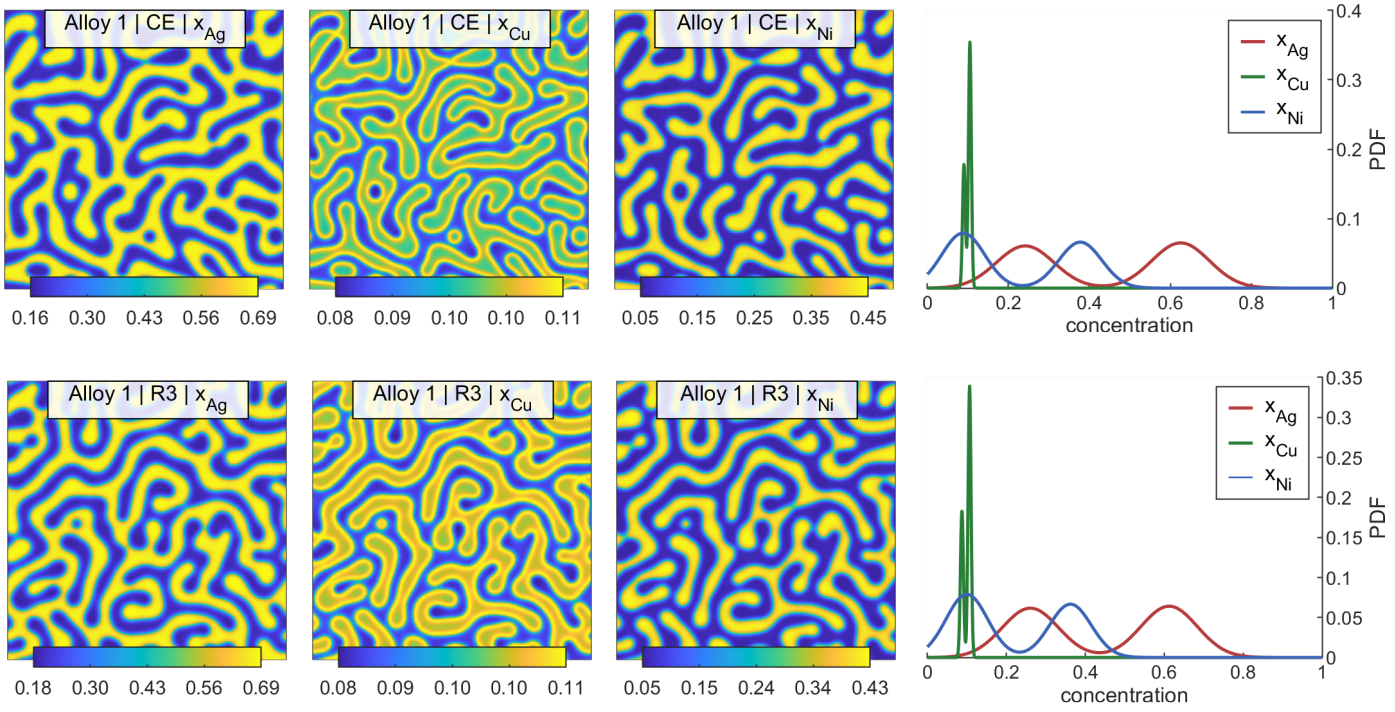
LIQUID #1	Moles	Mass
	0.531	54.709 g

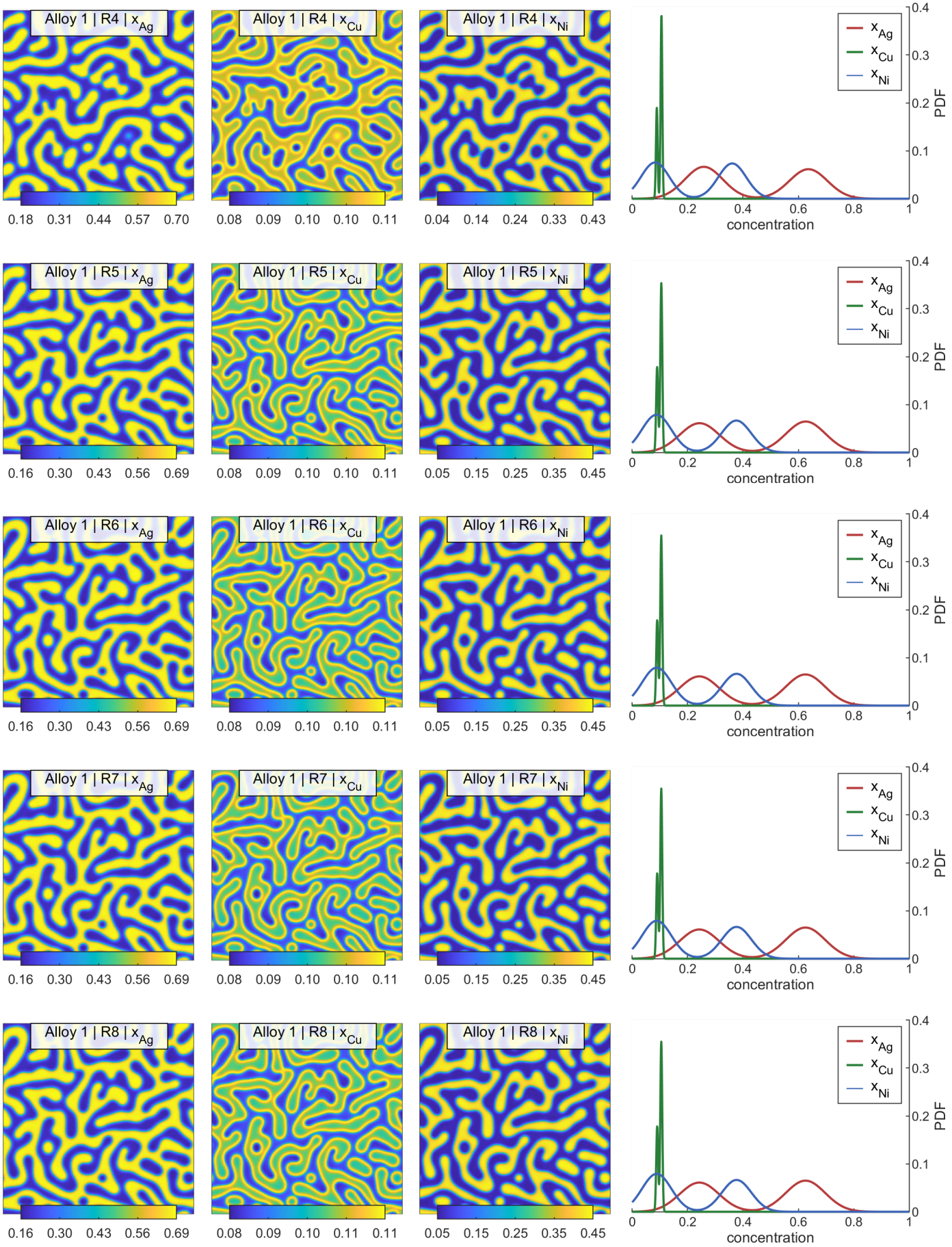
Component	Mole fraction	Mass fraction
Ag	0.661	0.691
Cu	0.094	0.058
Ni	0.056	0.032
Sn	0.191	0.219

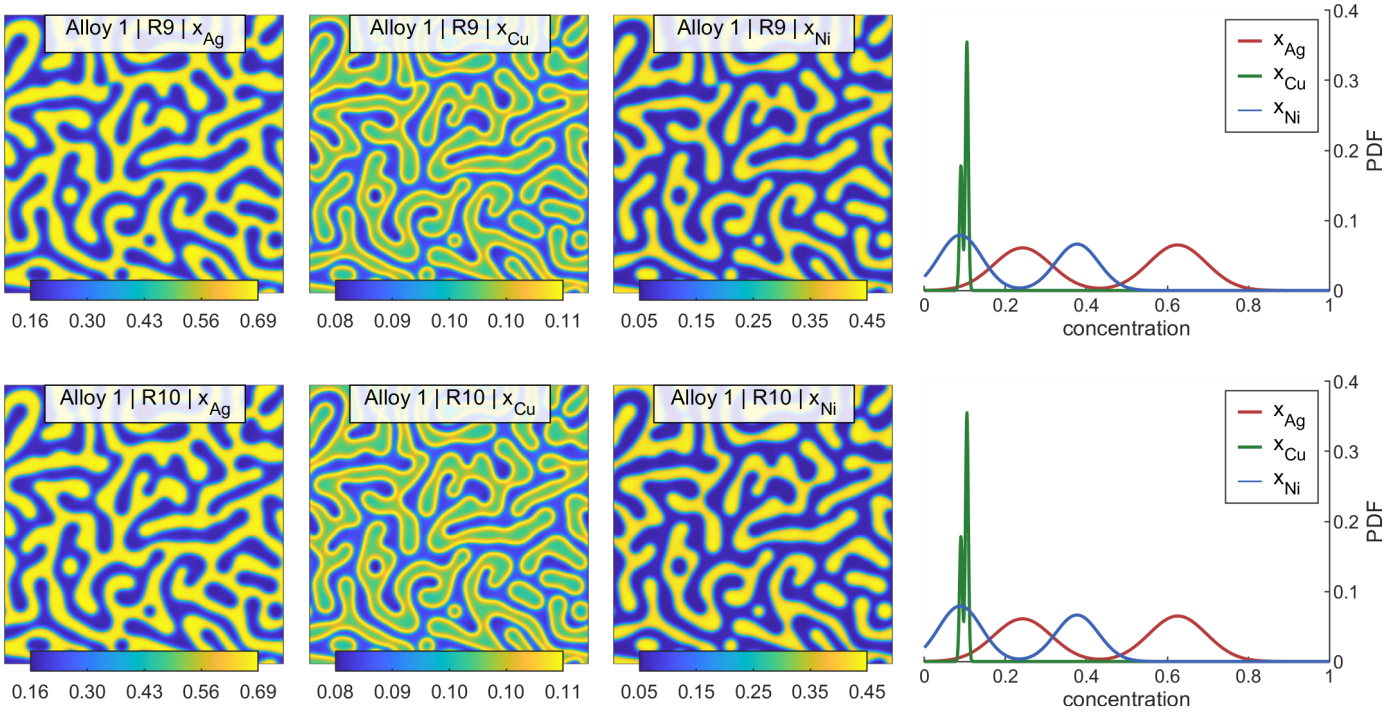
LIQUID #2	Moles	Mass
	0.469	40.511 g

Component	Mole fraction	Mass fraction
Ag	0.191	0.238
Cu	0.107	0.079
Ni	0.406	0.276
Sn	0.296	0.407

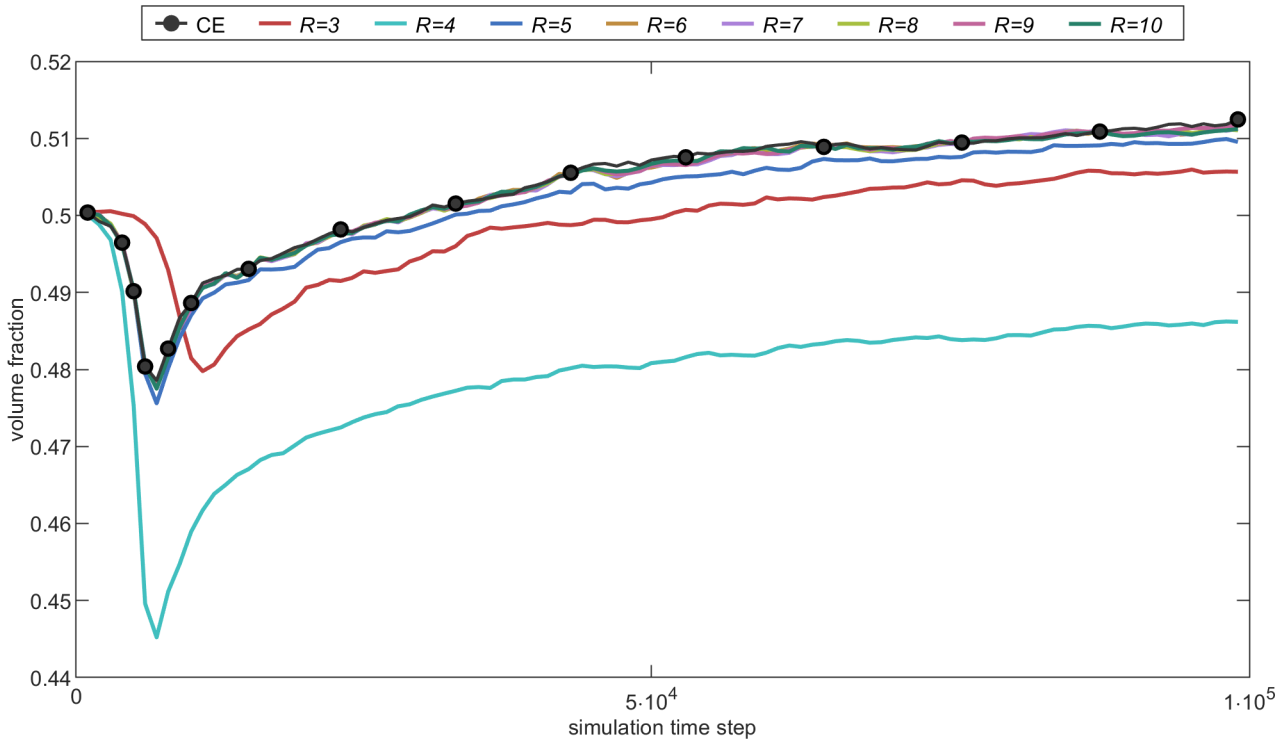
2D Microstructures




Alloy 1 | R4 | x_Ag
0.18 0.31 0.44 0.57 0.70
Alloy 1 | R4 | x_Cu
0.08 0.09 0.10 0.10 0.11
Alloy 1 | R4 | x_Ni
0.04 0.14 0.24 0.33 0.43
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 1 | R5 | x_Ag
0.16 0.30 0.43 0.56 0.69
Alloy 1 | R5 | x_Cu
0.08 0.09 0.10 0.10 0.11
Alloy 1 | R5 | x_Ni
0.05 0.15 0.25 0.35 0.45
Alloy 1 | R6 | x_Ag
0.16 0.30 0.43 0.56 0.69
Alloy 1 | R6 | x_Cu
0.08 0.09 0.10 0.10 0.11
Alloy 1 | R6 | x_Ni
0.05 0.15 0.25 0.35 0.45
Alloy 1 | R7 | x_Ag
0.16 0.30 0.43 0.56 0.69
Alloy 1 | R7 | x_Cu
0.08 0.09 0.10 0.10 0.11
Alloy 1 | R7 | x_Ni
0.05 0.15 0.25 0.35 0.45
Alloy 1 | R8 | x_Ag
0.16 0.30 0.43 0.56 0.69
Alloy 1 | R8 | x_Cu
0.08 0.09 0.10 0.11 0.11
Alloy 1 | R8 | x_Ni
0.05 0.15 0.25 0.35 0.45



Volume fraction



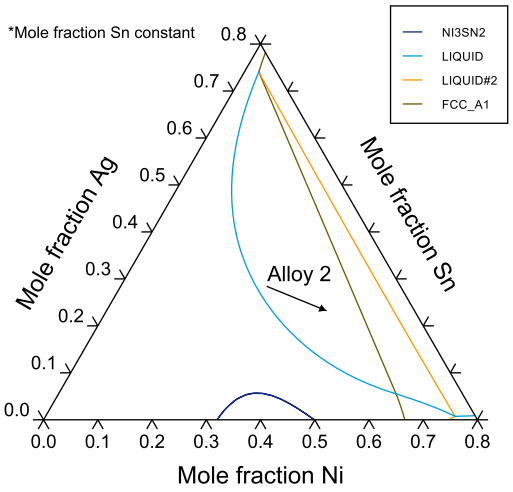
We note that, for all alloys, the equilibrium volume fraction of the phase, as computed with the Thermo-Calc software and COST531 database, is not yet reached in the simulations at time step 10^5. We verified that, for longer simulation times, the volume fractions in the simulations approach the equilibrium values calculated with the Thermo-Calc software. However, for the validation of the TTMs with different rank values in this study, we found it more useful to focus on the early evolution, where the structure and phase compositions are relatively far from thermodynamic equilibrium. Deviations on the thermodynamic quantities provided to the phase-field model will have more obvious effects on the simulation results in this initial regime, than when close to thermodynamic equilibrium.

Alloy 2

System definition

Component	Mole Fraction	Mass fraction
Ag	0.23	0.305
Cu	0.2	0.156
Ni	0.40	0.289
Sn	0.17	0.248

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

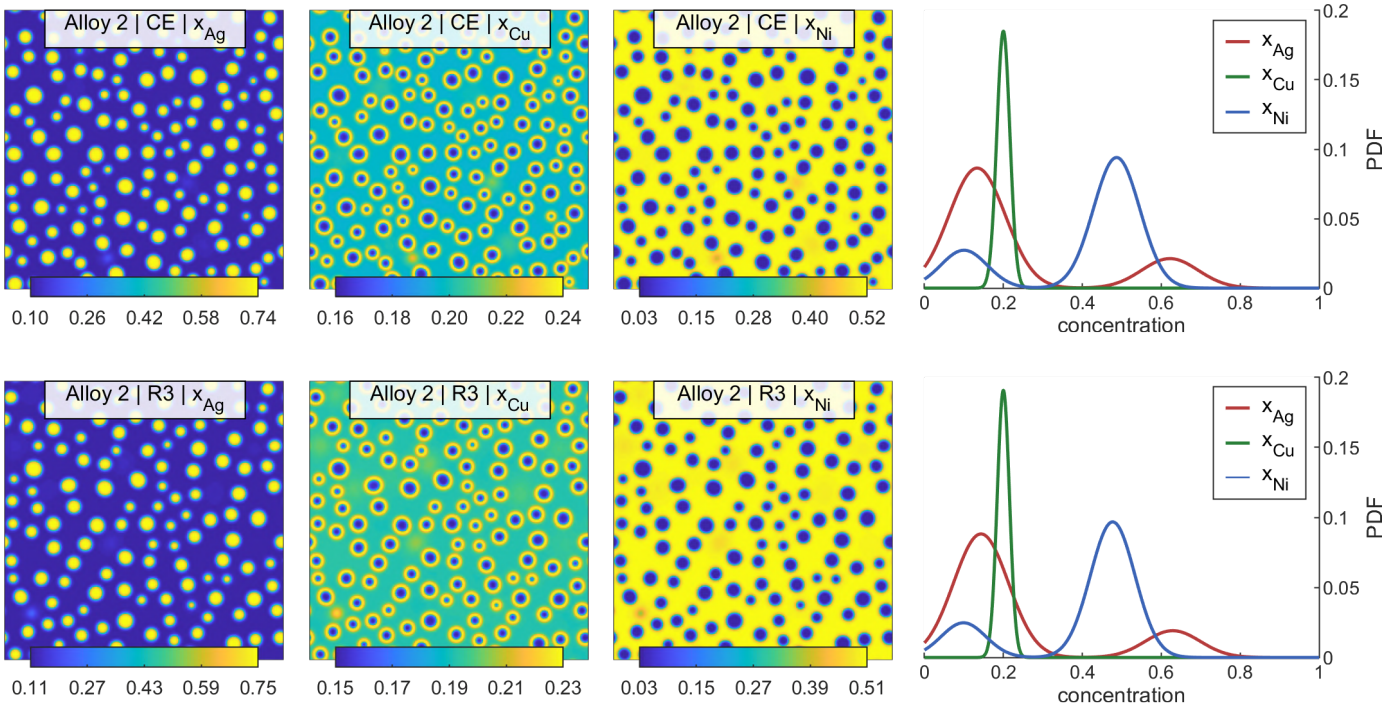
LIQUID #1	Moles	Mass
	0.218	21.585 g

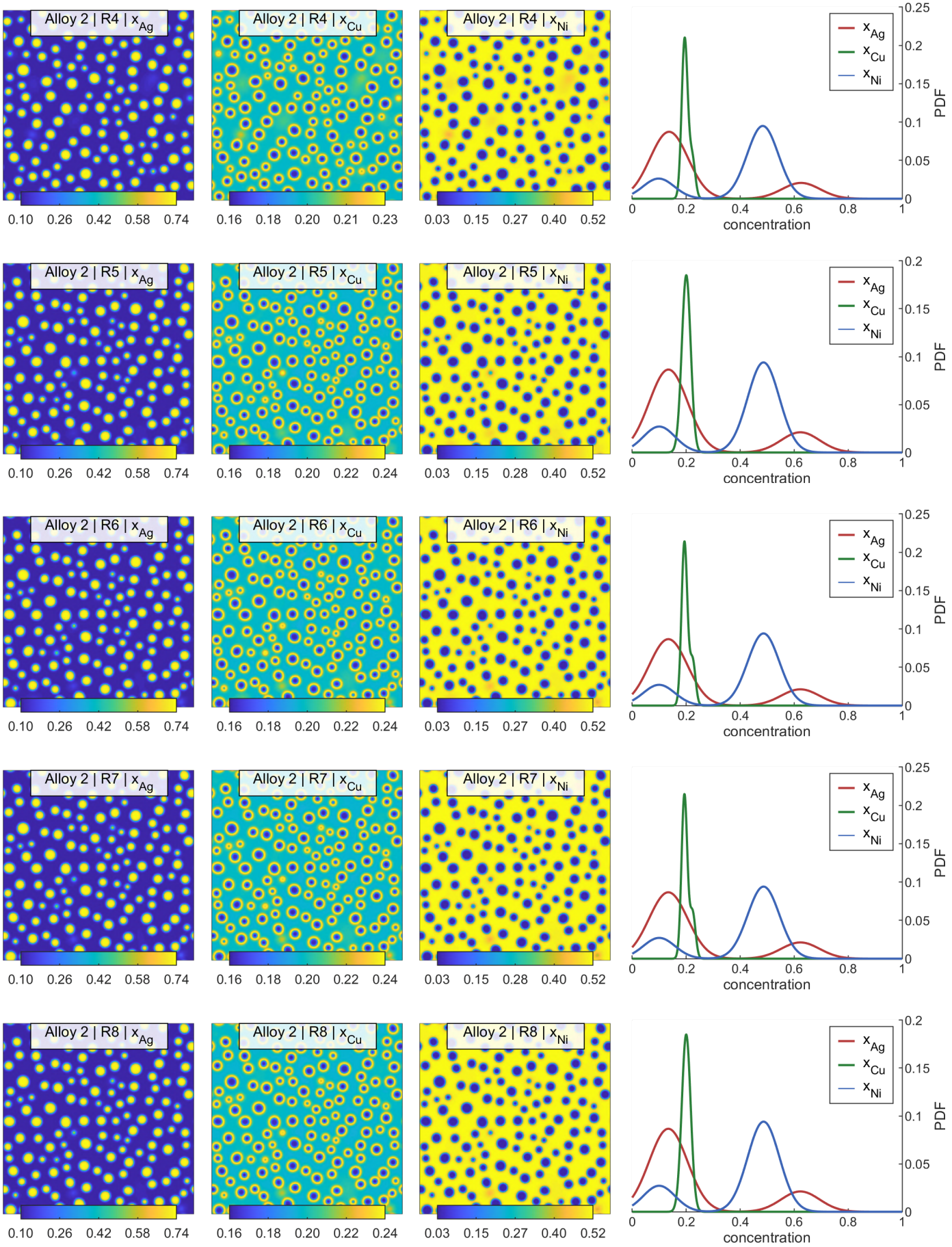
Component	Mole fraction	Mass fraction
Ag	0.694	0.755
Cu	0.179	0.115
Ni	0.036	0.021
Sn	0.091	0.108

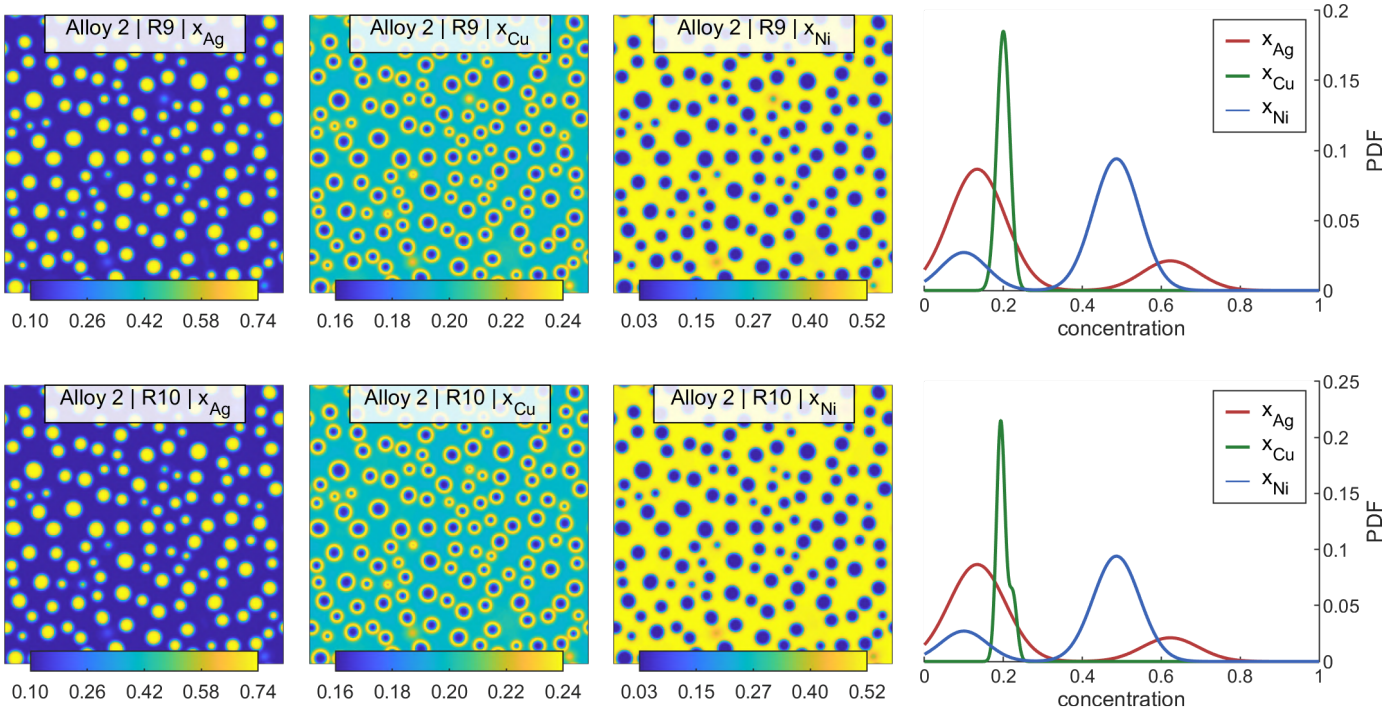
LIQUID #2	Moles	Mass
	0.782	59.591 g

Component	Mole fraction	Mass fraction
Ag	0.101	0.143
Cu	0.206	0.171
Ni	0.501	0.387
Sn	0.192	0.299

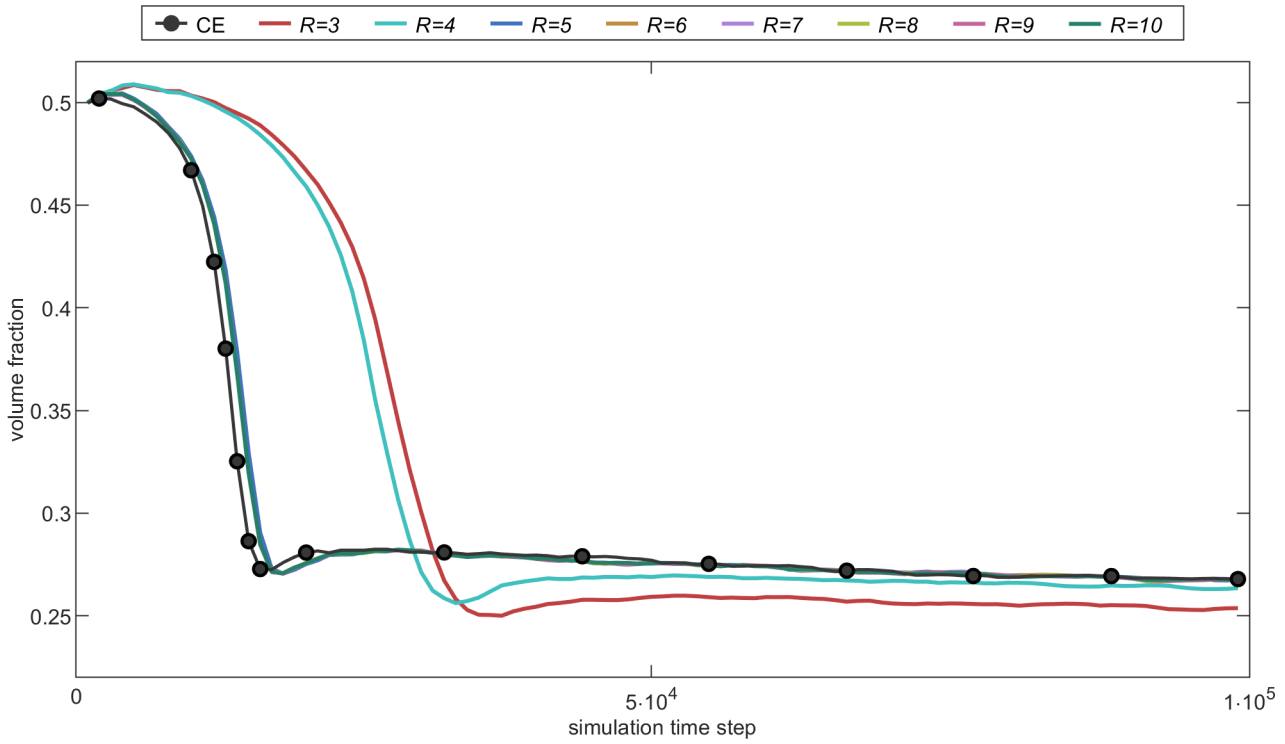
2D Microstructures




Alloy 2 | R4 | x_Ag
0.10 0.26 0.42 0.58 0.74
Alloy 2 | R4 | x_Cu
0.16 0.18 0.20 0.21 0.23
Alloy 2 | R4 | x_Ni
0.03 0.15 0.27 0.40 0.52
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 2 | R5 | x_Ag
0.10 0.26 0.42 0.58 0.74
Alloy 2 | R5 | x_Cu
0.16 0.18 0.20 0.22 0.24
Alloy 2 | R5 | x_Ni
0.03 0.15 0.28 0.40 0.52
Alloy 2 | R6 | x_Ag
0.10 0.26 0.42 0.58 0.74
Alloy 2 | R6 | x_Cu
0.16 0.18 0.20 0.22 0.24
Alloy 2 | R6 | x_Ni
0.03 0.15 0.27 0.40 0.52
Alloy 2 | R7 | x_Ag
0.10 0.26 0.42 0.58 0.74
Alloy 2 | R7 | x_Cu
0.16 0.18 0.20 0.22 0.24
Alloy 2 | R7 | x_Ni
0.03 0.15 0.28 0.40 0.52
Alloy 2 | R8 | x_Ag
0.10 0.26 0.42 0.58 0.74
Alloy 2 | R8 | x_Cu
0.16 0.18 0.20 0.22 0.24
Alloy 2 | R8 | x_Ni
0.03 0.15 0.27 0.40 0.52



Volume fraction

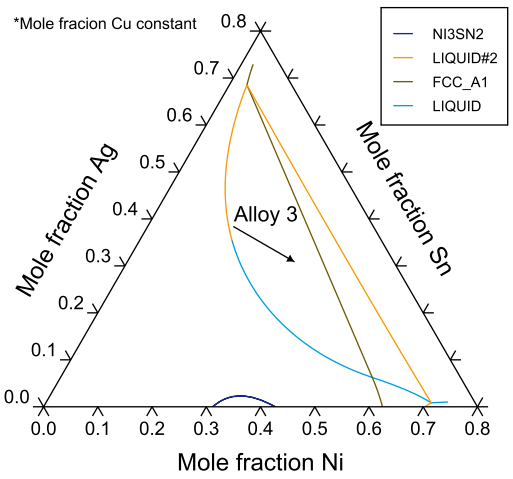


Alloy 3

System definition

Component	Mole Fraction	Mass fraction
Ag	0.3	0.387
Cu	0.25	0.189
Ni	0.3	0.211
Sn	0.15	0.213

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

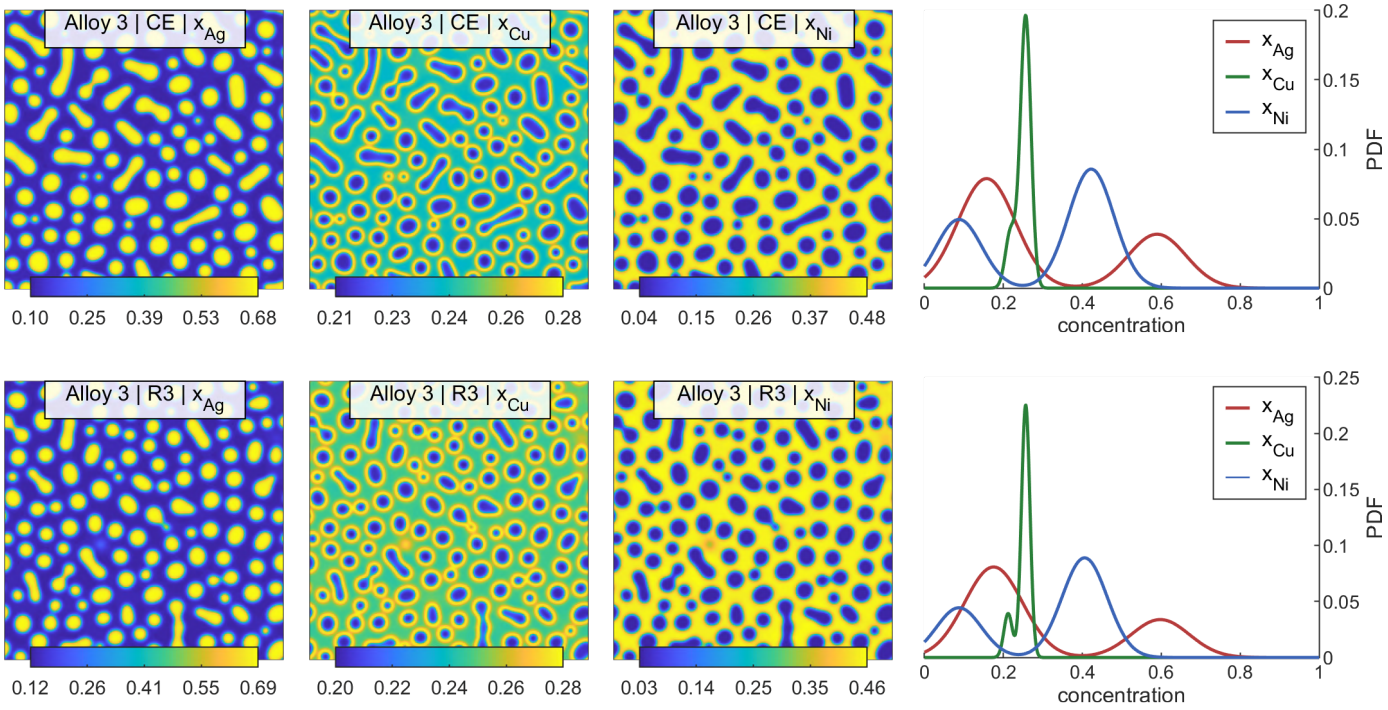
LIQUID #1	Moles	Mass
	0.358	34.461 g

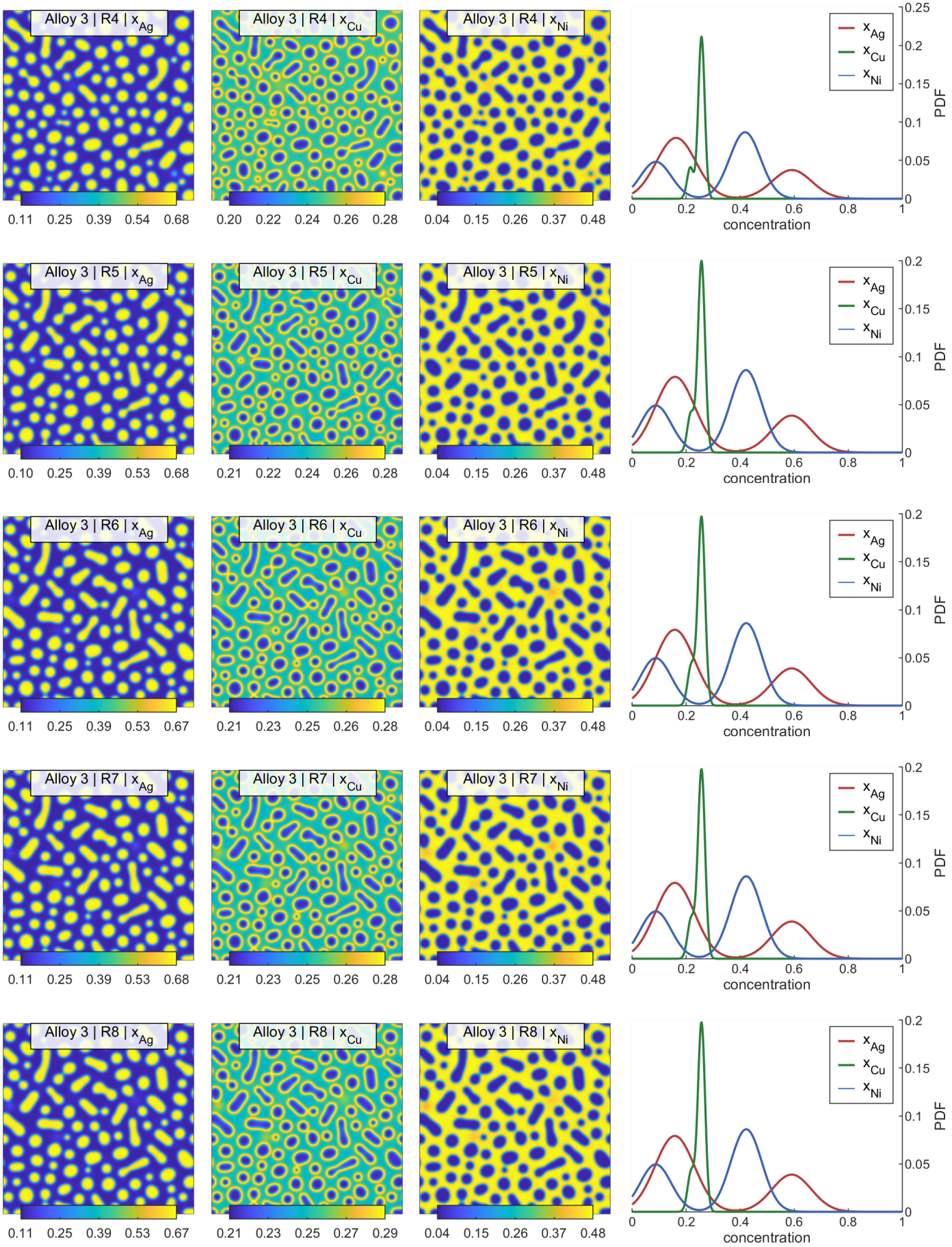
Component	Mole fraction	Mass fraction
Ag	0.629	0.704
Cu	0.232	0.153
Ni	0.045	0.027
Sn	0.094	0.116

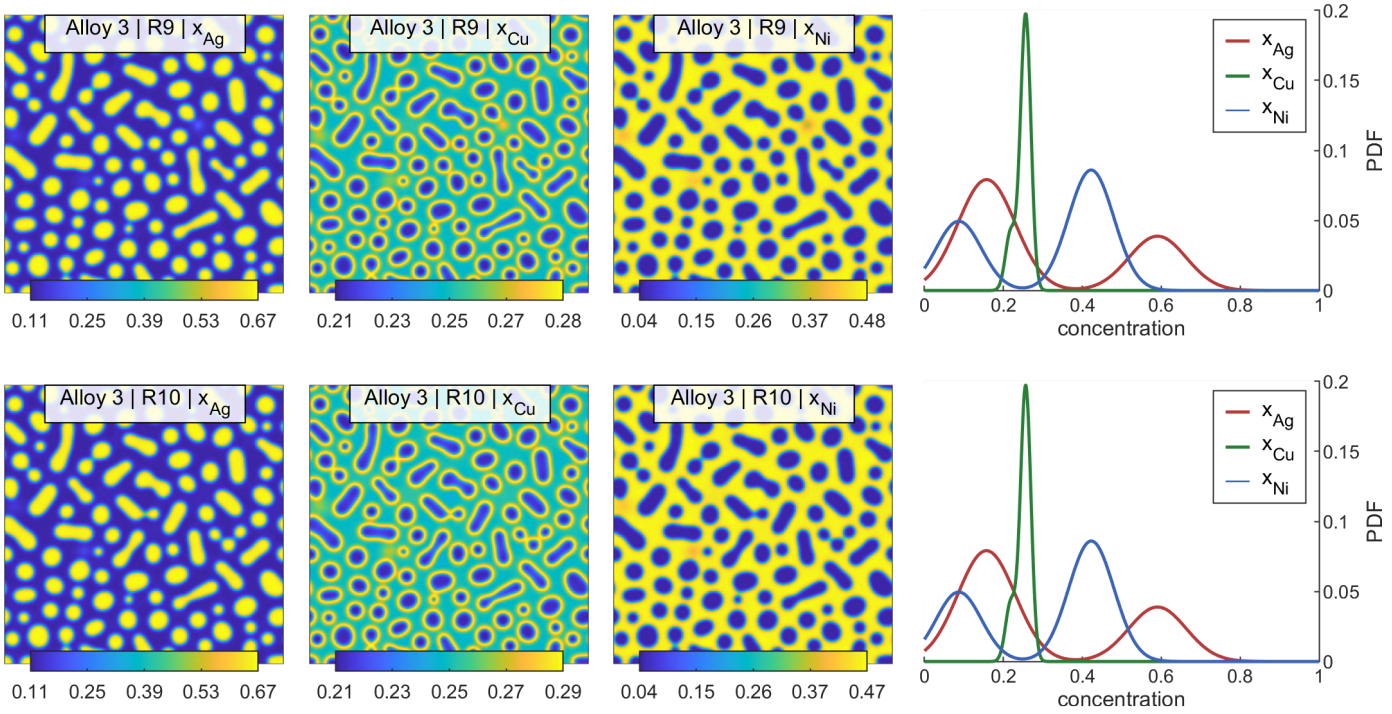
LIQUID #2	Moles	Mass
	0.642	49.201 g

Component	Mole fraction	Mass fraction
Ag	0.117	0.165
Cu	0.261	0.216
Ni	0.442	0.338
Sn	0.181	0.281

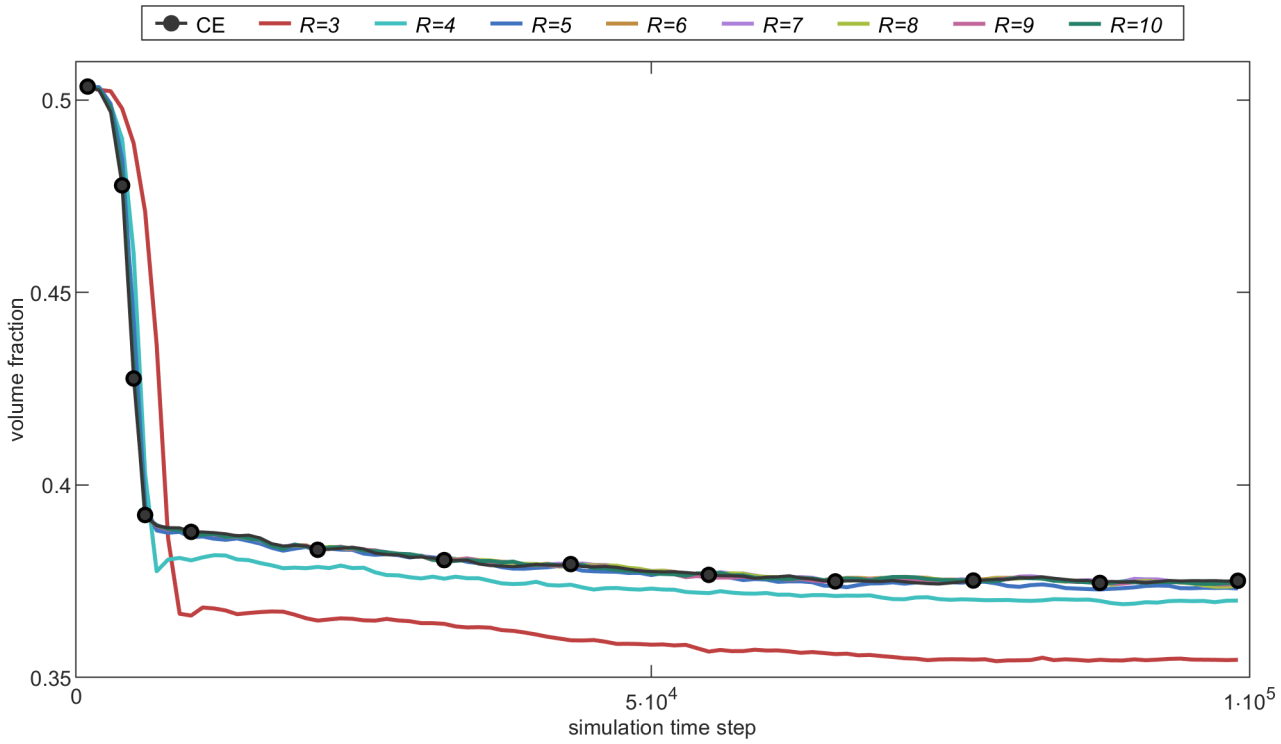
2D Microstructures




Alloy 3 | R4 | x_Ag
0.11 0.25 0.39 0.54 0.68
Alloy 3 | R4 | x_Cu
0.20 0.22 0.24 0.26 0.28
Alloy 3 | R4 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 3 | R5 | x_Ag
0.10 0.25 0.39 0.53 0.68
Alloy 3 | R5 | x_Cu
0.21 0.22 0.24 0.26 0.28
Alloy 3 | R5 | x_Ni
0.04 0.15 0.26 0.37 0.48
Alloy 3 | R6 | x_Ag
0.11 0.25 0.39 0.53 0.67
Alloy 3 | R6 | x_Cu
0.21 0.23 0.25 0.26 0.28
Alloy 3 | R6 | x_Ni
0.04 0.15 0.26 0.37 0.48
Alloy 3 | R7 | x_Ag
0.11 0.25 0.39 0.53 0.68
Alloy 3 | R7 | x_Cu
0.21 0.23 0.25 0.26 0.28
Alloy 3 | R7 | x_Ni
0.04 0.15 0.26 0.37 0.48
Alloy 3 | R8 | x_Ag
0.11 0.25 0.39 0.53 0.67
Alloy 3 | R8 | x_Cu
0.21 0.23 0.25 0.27 0.29
Alloy 3 | R8 | x_Ni
0.04 0.15 0.26 0.37 0.48



Volume fraction

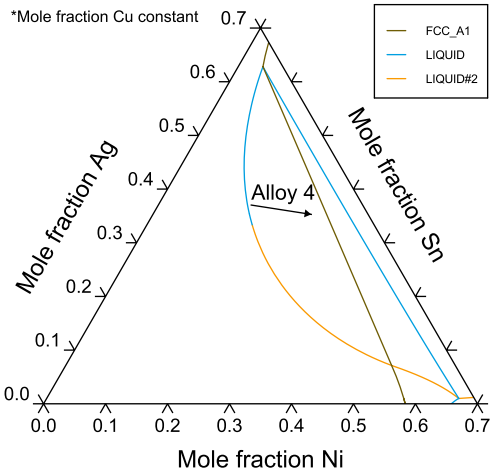


Alloy 4

System definition

Component	Mole Fraction	Mass fraction
Ag	0.35	0.453
Cu	0.3	0.228
Ni	0.25	0.176
Sn	0.1	0.142

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

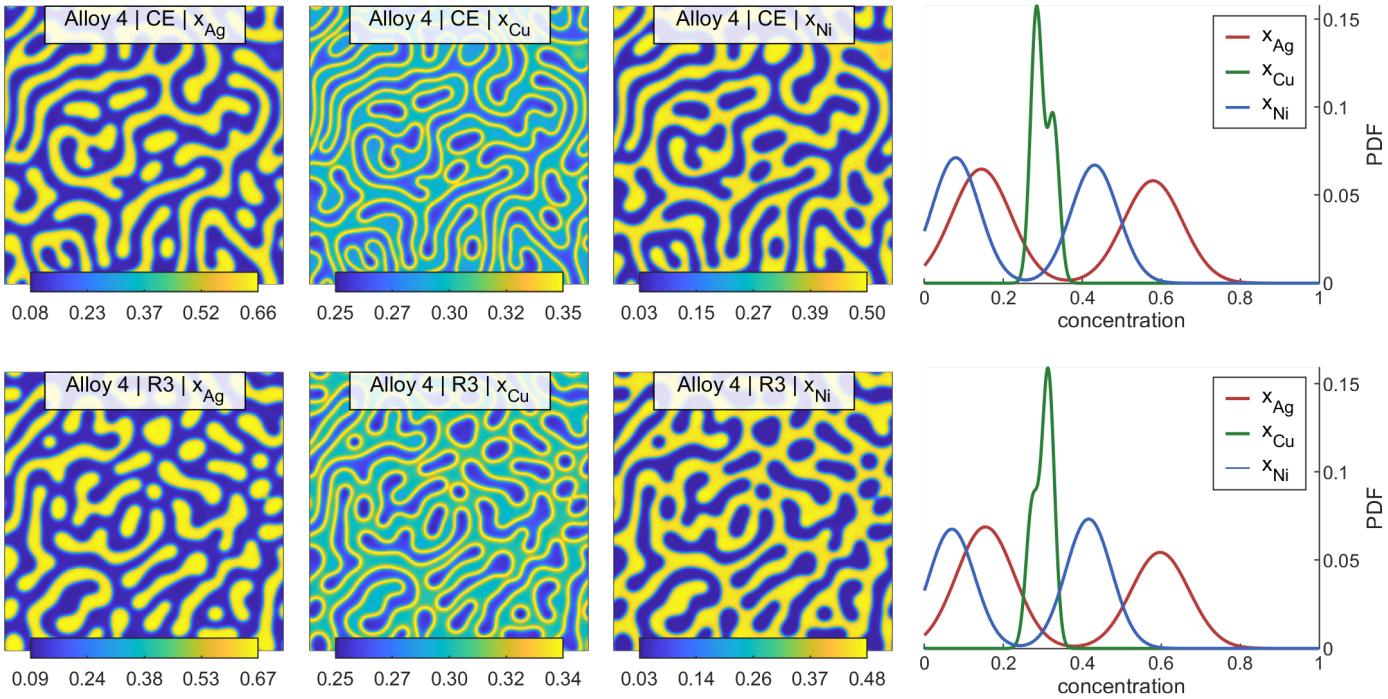
LIQUID #1	Moles	Mass
	0.492	46.099 g

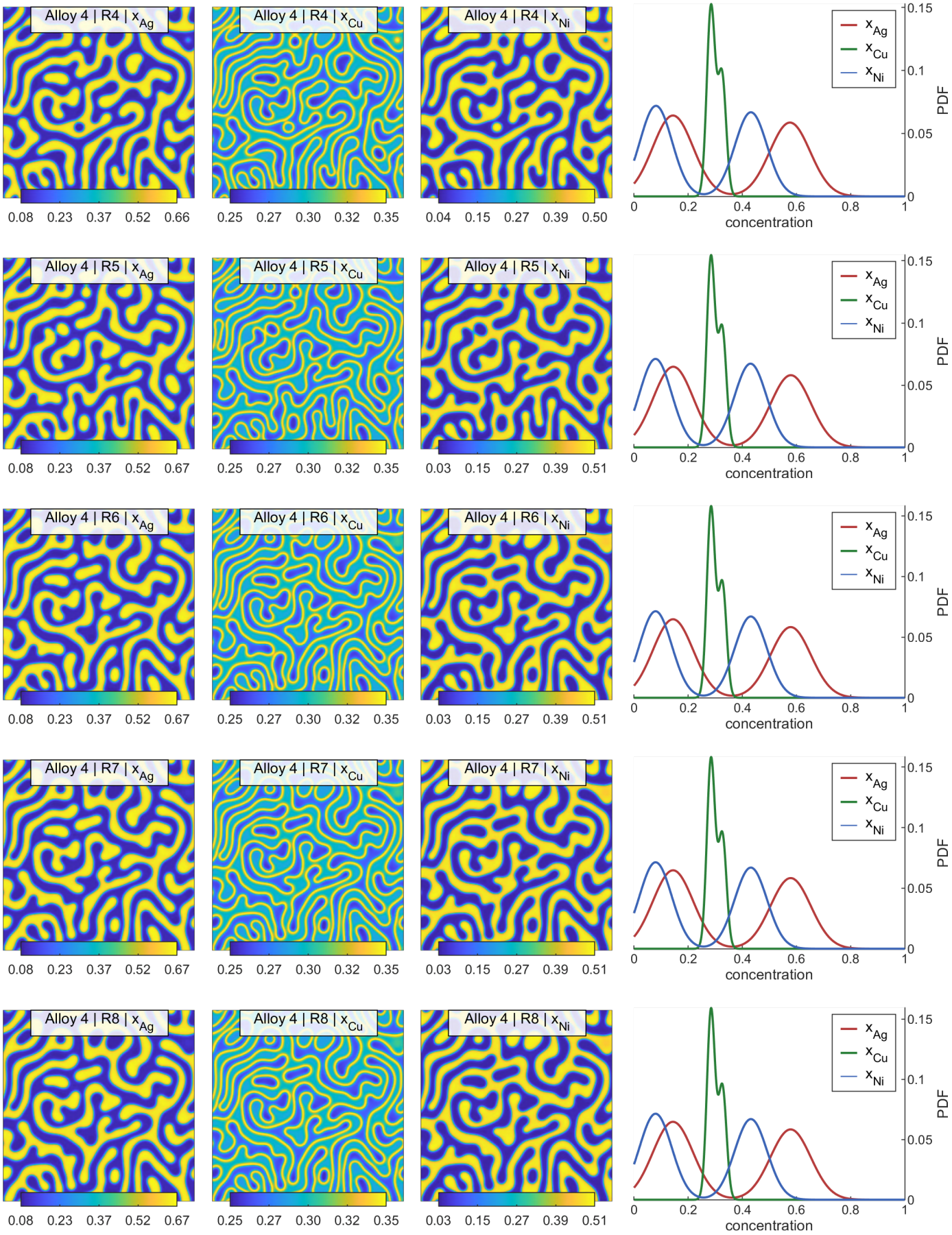
Component	Mole fraction	Mass fraction
Ag	0.607	0.699
Cu	0.286	0.194
Ni	0.045	0.028
Sn	0.062	0.078

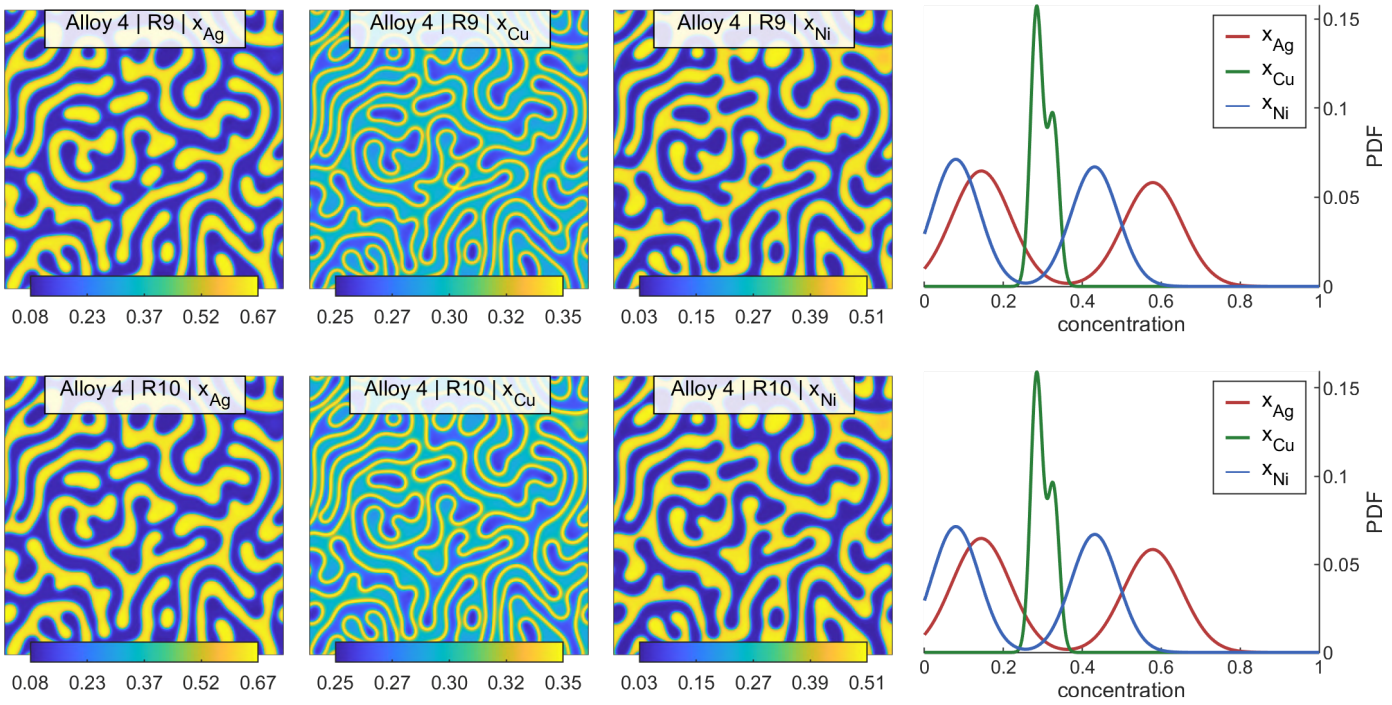
LIQUID #2	Moles	Mass
	0.507	37.0262 g

Component	Mole fraction	Mass fraction
Ag	0.101	0.148
Cu	0.314	0.271
Ni	0.448	0.358
Sn	0.137	0.221

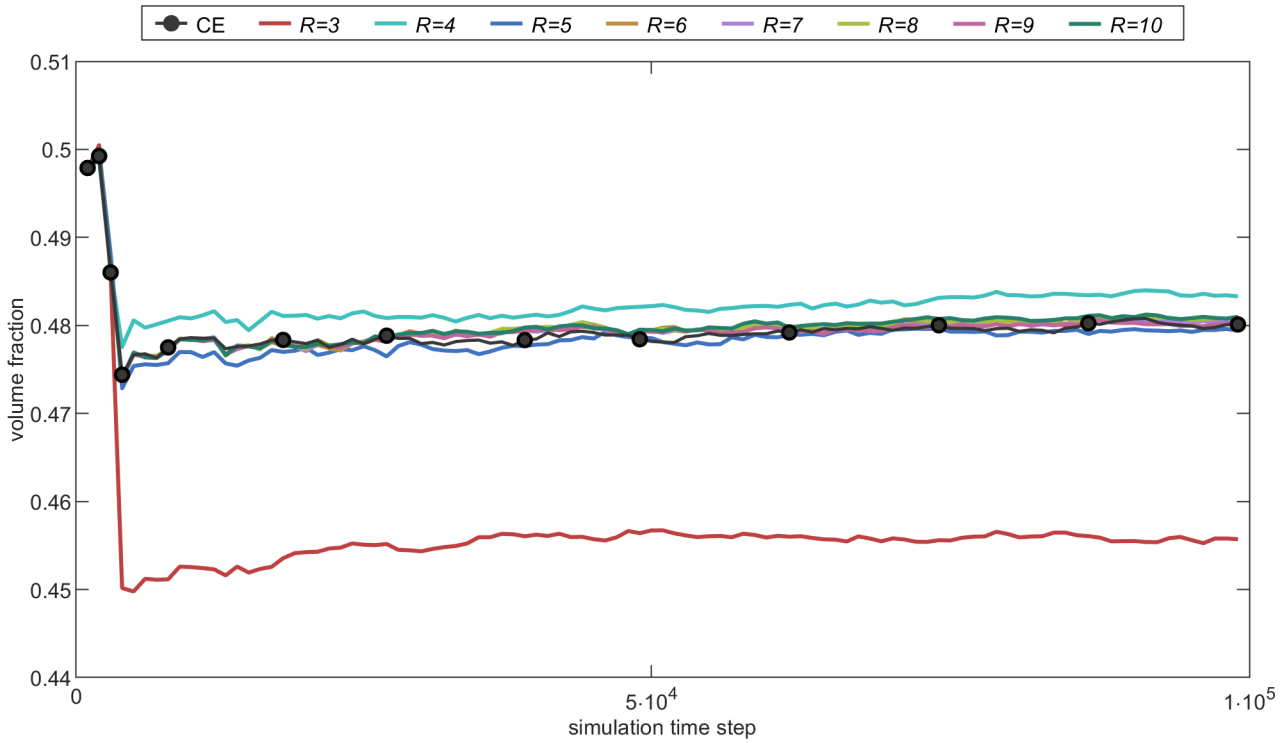
2D Microstructures




Alloy 4 | R4 | x_Ag
0.08 0.23 0.37 0.52 0.66
Alloy 4 | R4 | x_Cu
0.25 0.27 0.30 0.32 0.35
Alloy 4 | R4 | x_Ni
0.04 0.15 0.27 0.39 0.50
x_Ag
x_Cu
x_Ni
PDF
0 0.05 0.1 0.15
0 0.2 0.4 0.6 0.8 1
concentration
Alloy 4 | R5 | x_Ag
0.08 0.23 0.37 0.52 0.67
Alloy 4 | R5 | x_Cu
0.25 0.27 0.30 0.32 0.35
Alloy 4 | R5 | x_Ni
0.03 0.15 0.27 0.39 0.51
x_Ag
x_Cu
x_Ni
PDF
0 0.05 0.1 0.15
0 0.2 0.4 0.6 0.8 1
concentration
Alloy 4 | R6 | x_Ag
0.08 0.23 0.37 0.52 0.67
Alloy 4 | R6 | x_Cu
0.25 0.27 0.30 0.32 0.35
Alloy 4 | R6 | x_Ni
0.03 0.15 0.27 0.39 0.51
x_Ag
x_Cu
x_Ni
PDF
0 0.05 0.1 0.15
0 0.2 0.4 0.6 0.8 1
concentration
Alloy 4 | R7 | x_Ag
0.08 0.23 0.37 0.52 0.67
Alloy 4 | R7 | x_Cu
0.25 0.27 0.30 0.32 0.35
Alloy 4 | R7 | x_Ni
0.03 0.15 0.27 0.39 0.51
x_Ag
x_Cu
x_Ni
PDF
0 0.05 0.1 0.15
0 0.2 0.4 0.6 0.8 1
concentration
Alloy 4 | R8 | x_Ag
0.08 0.23 0.37 0.52 0.67
Alloy 4 | R8 | x_Cu
0.25 0.27 0.30 0.32 0.35
Alloy 4 | R8 | x_Ni
0.03 0.15 0.27 0.39 0.51
x_Ag
x_Cu
x_Ni
PDF
0 0.05 0.1 0.15
0 0.2 0.4 0.6 0.8 1
concentration



Volume fraction

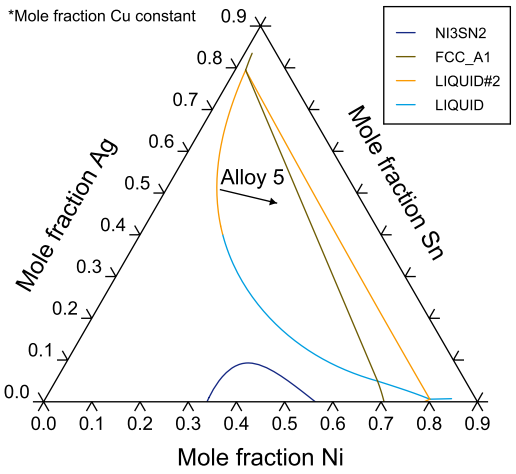


Alloy 5

System definition

Component	Mole Fraction	Mass fraction
Ag	0.24	0.325
Cu	0.15	0.119
Ni	0.47	0.346
Sn	0.14	0.209

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

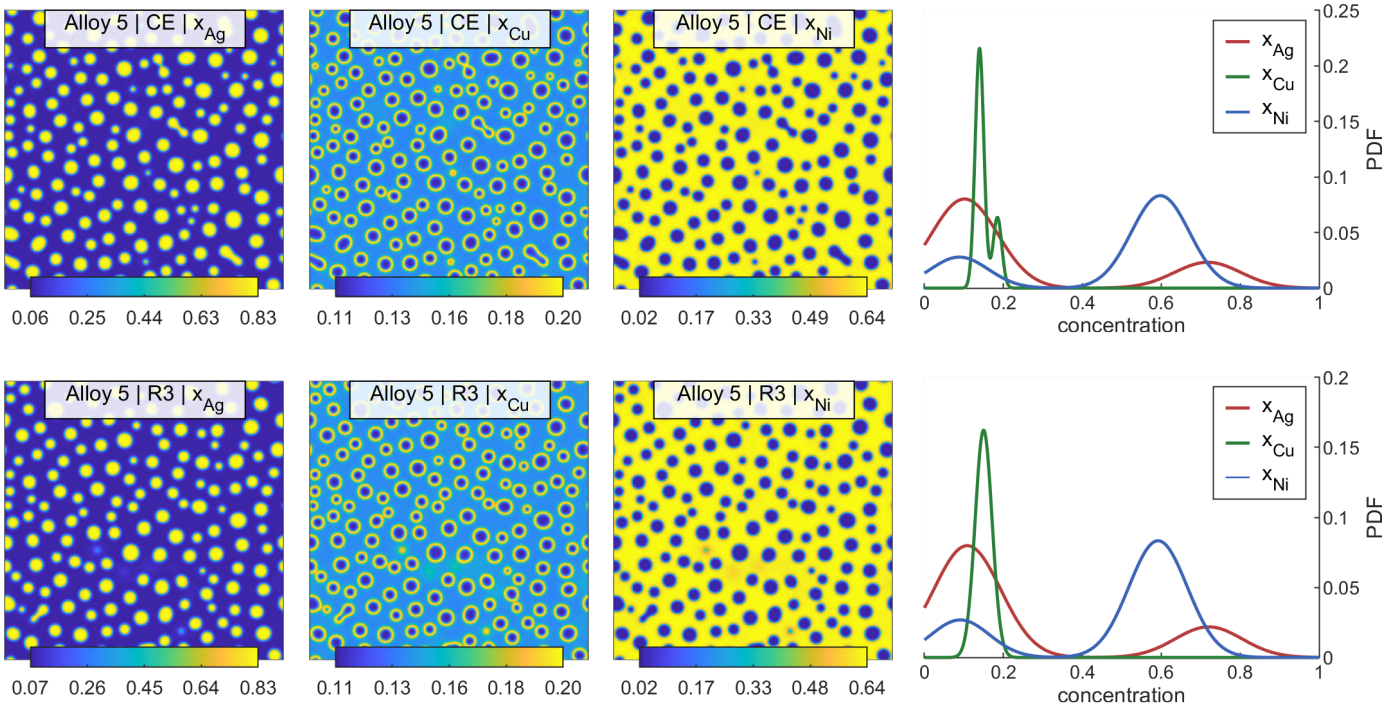
LIQUID #1	Moles	Mass
	0.757	55.041 g

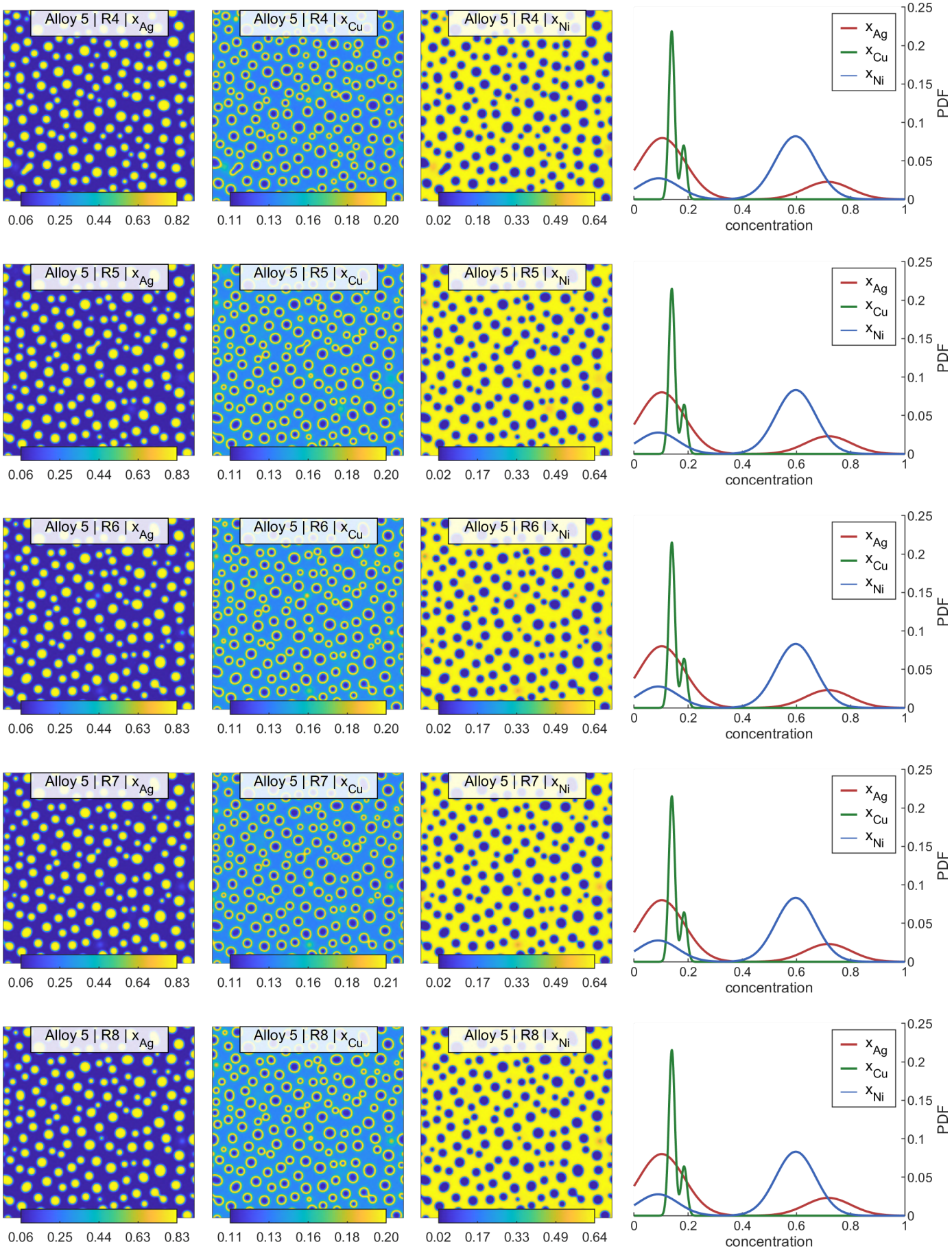
Component	Mole fraction	Mass fraction
Ag	0.066	0.098
Cu	0.166	0.272
Ni	0.155	0.135
Sn	0.613	0.495

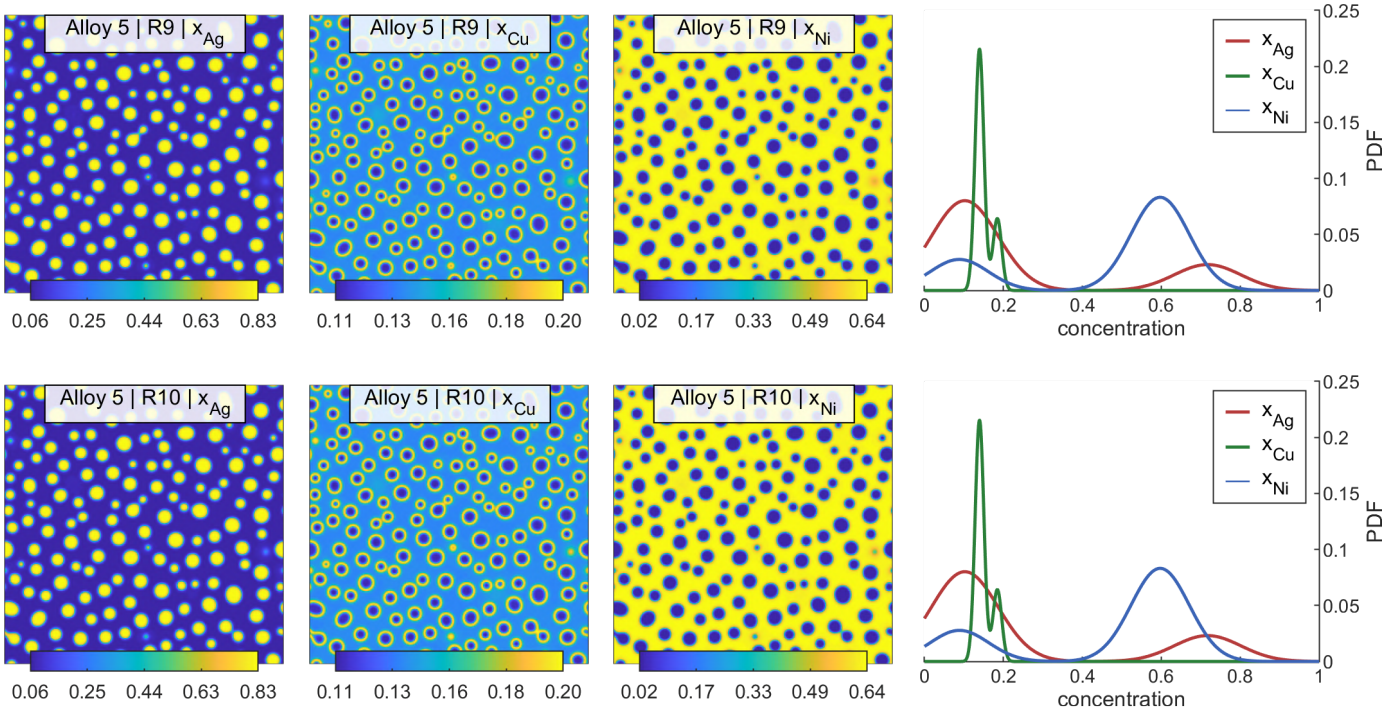
LIQUID #2	Moles	Mass
	0.243	24.584 g

Component	Mole fraction	Mass fraction
Ag	0.783	0.834
Cu	0.136	0.085
Ni	0.024	0.014
Sn	0.057	0.067

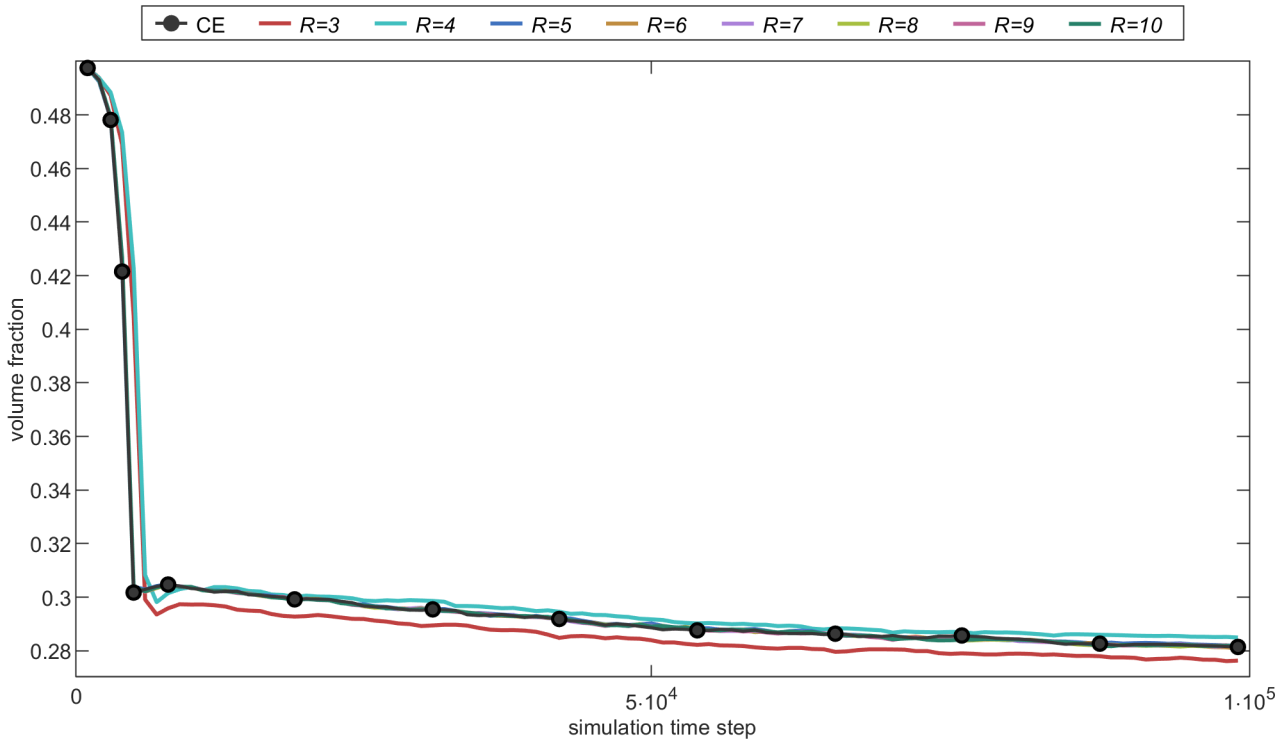
2D Microstructures




Alloy 5 | R4 | x_Ag
0.06 0.25 0.44 0.63 0.82
Alloy 5 | R4 | x_Cu
0.11 0.13 0.16 0.18 0.20
Alloy 5 | R4 | x_Ni
0.02 0.18 0.33 0.49 0.64
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 5 | R5 | x_Ag
0.06 0.25 0.44 0.63 0.83
Alloy 5 | R5 | x_Cu
0.11 0.13 0.16 0.18 0.20
Alloy 5 | R5 | x_Ni
0.02 0.17 0.33 0.49 0.64
Alloy 5 | R6 | x_Ag
0.06 0.25 0.44 0.63 0.83
Alloy 5 | R6 | x_Cu
0.11 0.13 0.16 0.18 0.20
Alloy 5 | R6 | x_Ni
0.02 0.17 0.33 0.49 0.64
Alloy 5 | R7 | x_Ag
0.06 0.25 0.44 0.64 0.83
Alloy 5 | R7 | x_Cu
0.11 0.13 0.16 0.18 0.21
Alloy 5 | R7 | x_Ni
0.02 0.17 0.33 0.49 0.64
Alloy 5 | R8 | x_Ag
0.06 0.25 0.44 0.63 0.83
Alloy 5 | R8 | x_Cu
0.11 0.13 0.16 0.18 0.20
Alloy 5 | R8 | x_Ni
0.02 0.17 0.33 0.49 0.64



Volume fraction

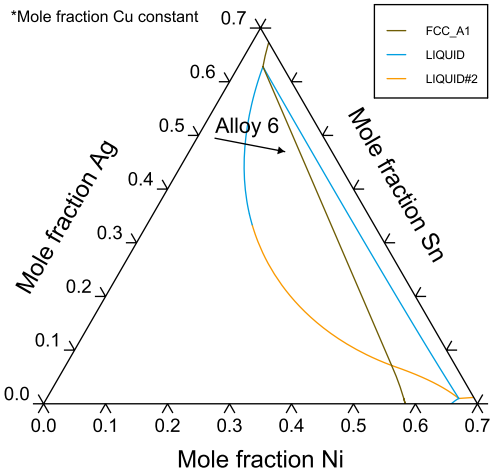


Alloy 6

System definition

Component	Mole Fraction	Mass fraction
Ag	0.46	0.563
Cu	0.3	0.216
Ni	0.15	0.099
Sn	0.09	0.122

Moles	1
Temperature	1400 K



Liquid phase equilibrium condition

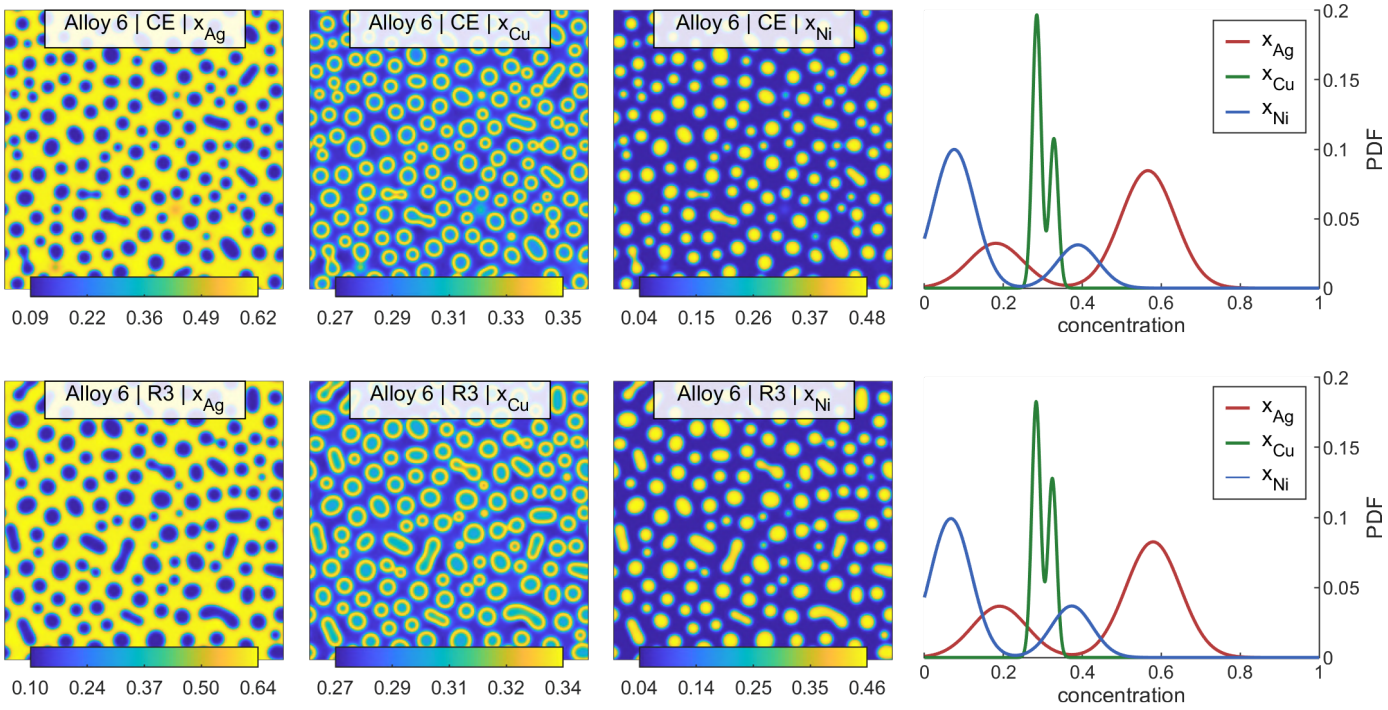
LIQUID #1	Moles	Mass
	0.732	68.267 g

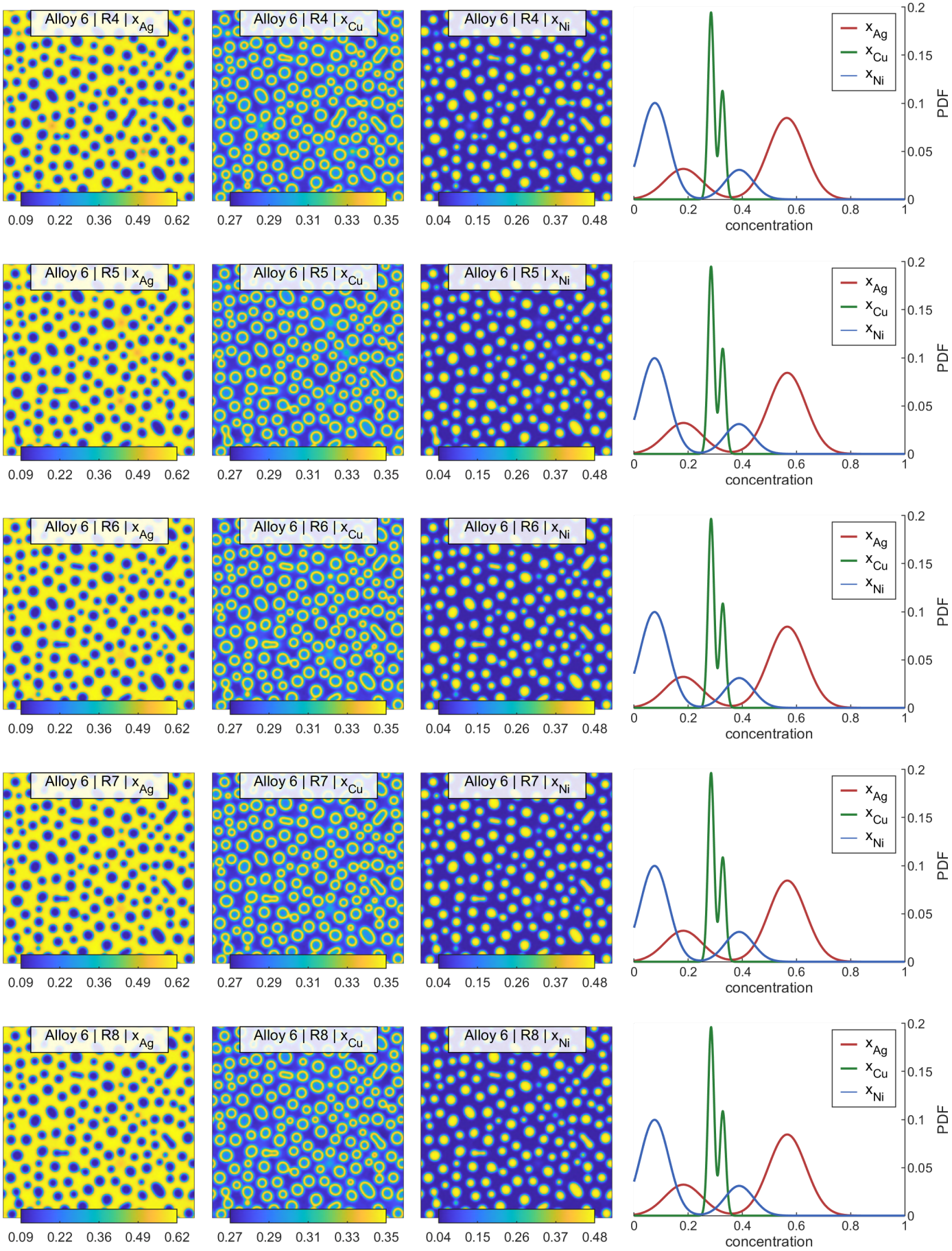
Component	Mole fraction	Mass fraction
Ag	0.588	0.679
Cu	0.292	0.199
Ni	0.049	0.031
Sn	0.071	0.089

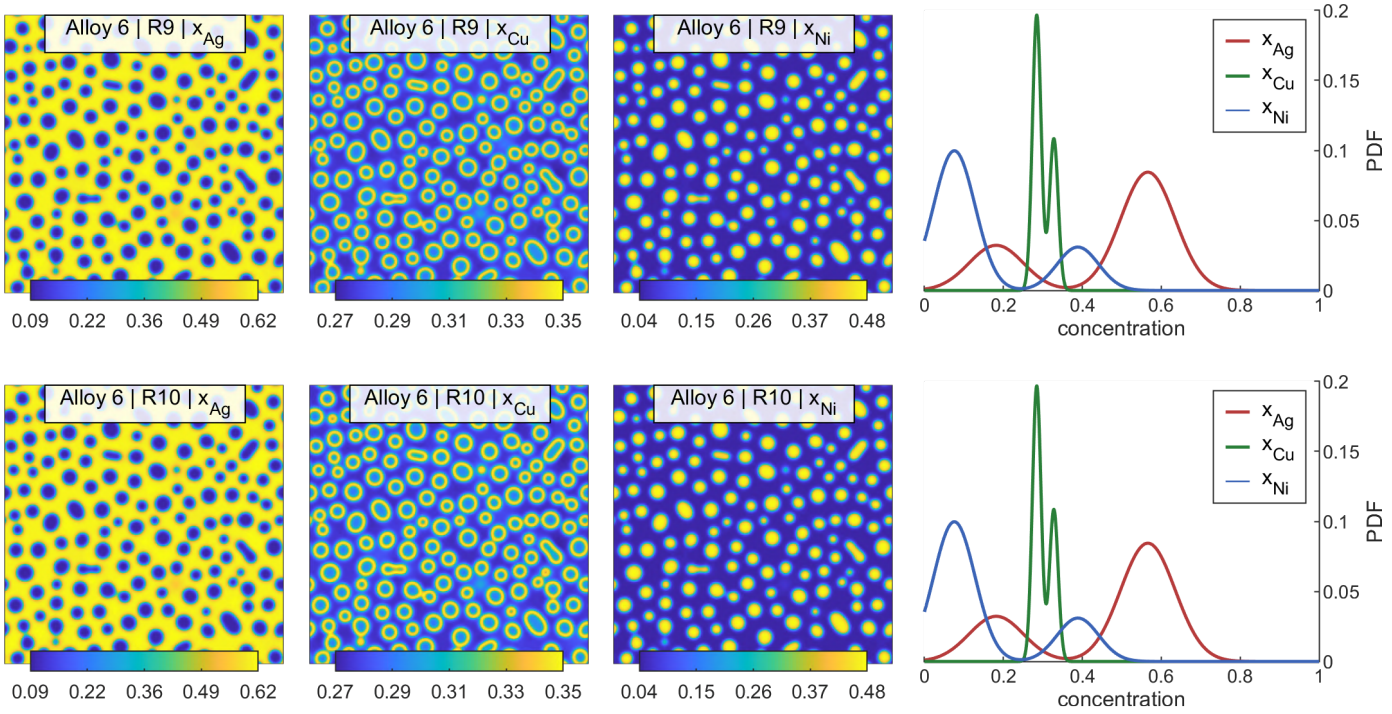
LIQUID #2	Moles	Mass
	0.268	19.904 g

Component	Mole fraction	Mass fraction
Ag	0.111	0.161
Cu	0.321	0.274
Ni	0.425	0.336
Sn	0.143	0.229

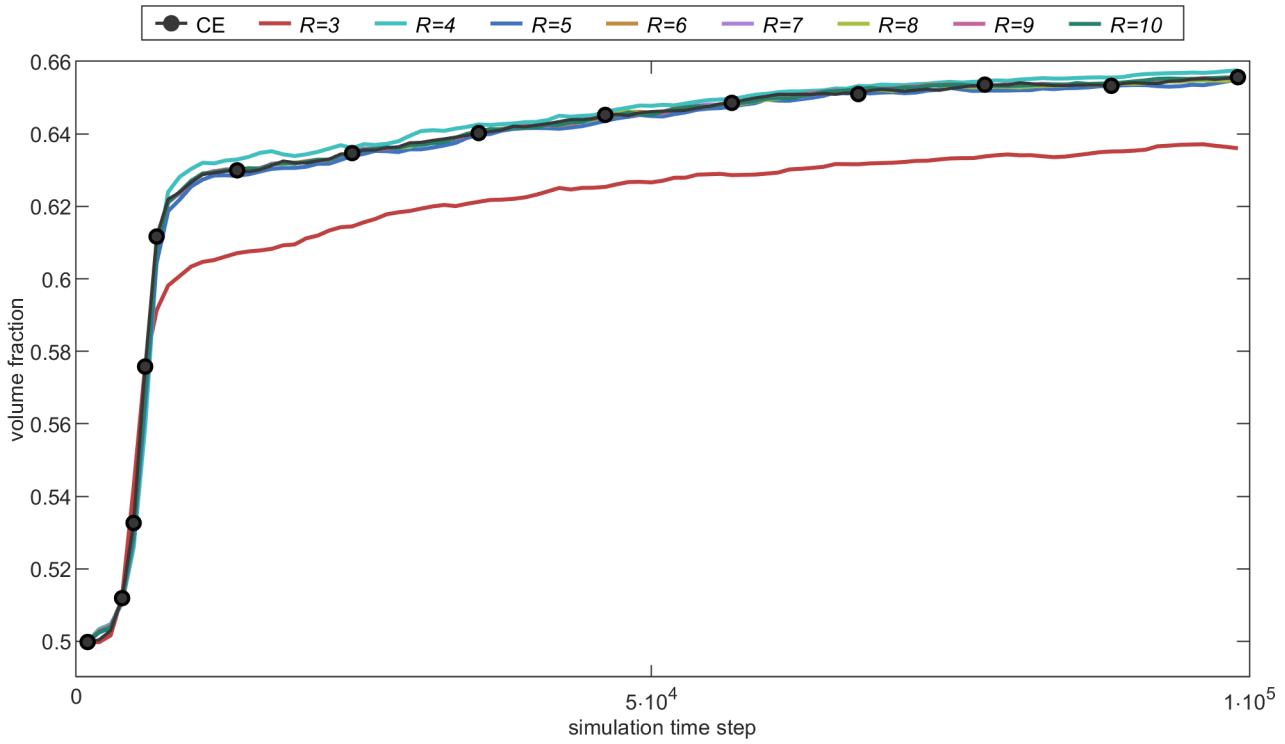
2D Microstructures




Alloy 6 | R4 | x_Ag
0.09 0.22 0.36 0.49 0.62
Alloy 6 | R4 | x_Cu
0.27 0.29 0.31 0.33 0.35
Alloy 6 | R4 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 6 | R5 | x_Ag
0.09 0.22 0.36 0.49 0.62
Alloy 6 | R5 | x_Cu
0.27 0.29 0.31 0.33 0.35
Alloy 6 | R5 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 6 | R6 | x_Ag
0.09 0.22 0.36 0.49 0.62
Alloy 6 | R6 | x_Cu
0.27 0.29 0.31 0.33 0.35
Alloy 6 | R6 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 6 | R7 | x_Ag
0.09 0.22 0.36 0.49 0.62
Alloy 6 | R7 | x_Cu
0.27 0.29 0.31 0.33 0.35
Alloy 6 | R7 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration
Alloy 6 | R8 | x_Ag
0.09 0.22 0.36 0.49 0.62
Alloy 6 | R8 | x_Cu
0.27 0.29 0.31 0.33 0.35
Alloy 6 | R8 | x_Ni
0.04 0.15 0.26 0.37 0.48
x_Ag
x_Cu
x_Ni
PDF
concentration

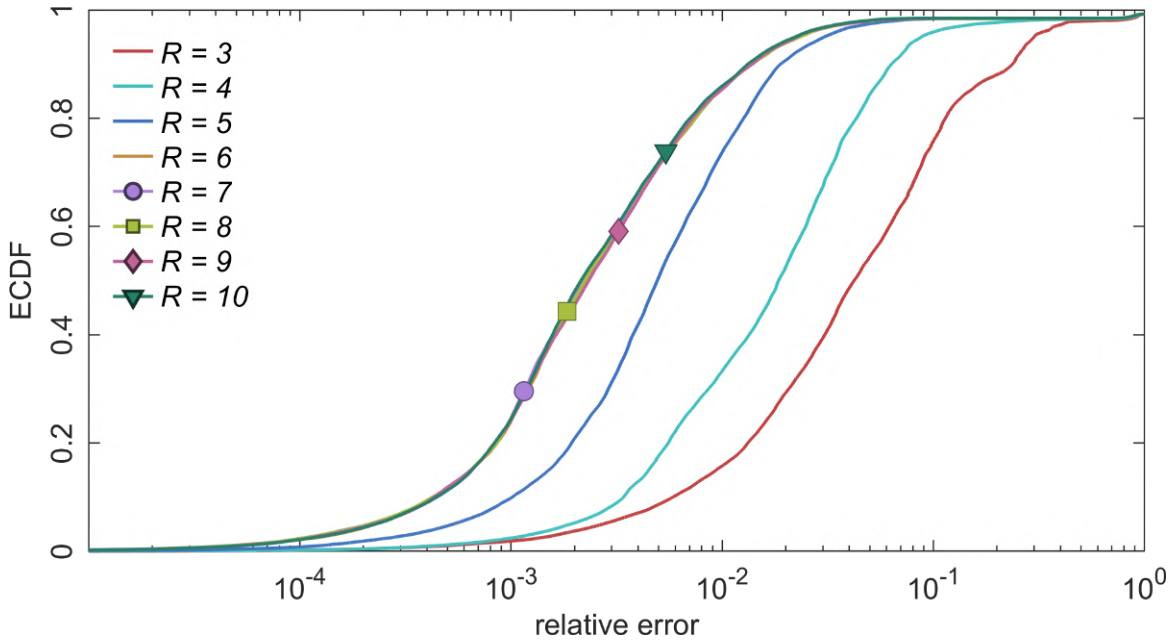


Volume fraction



1D simulations

For each of the six alloy compositions presented in the table at the beginning of the document, five 1D simulations are performed, giving a total of 270 1D simulations. In this case, simulations where the thermodynamic quantities are provided via a programming interface with the TC-Toolbox (Thermo-Calc 2017b), are taken as reference with which the simulations using TTMs with rank values $R = 3, \ldots, 10$ are compared to verify their accuracy. In the same way as for the 2D simulations, the Langer-Bar-On-Miller (LBM) method was used to characterise the molar fraction fields of the elements Ag, Cu and Ni for each of the simulations and at each time step. The empirical cumulative probability distribution function (ECDF) of the relative error on the amplitudes of the distributions is plotted in the following figure.



The conclusions obtained from the 1D simulation are not discussed in the main text since they are similar to those obtained from the 2D simulations. The ECDFs in the figure show that the accuracy of the simulations increases as the rank value of the TTM increases, up to $R = 6$, and no further improvement is observed for higher rank values.

The method in which thermodynamic quantities are provided via a programming interface showed to be extremely inefficient in our phase-field implementation. For this reason, it was not used for the 2D simulations. In the literature, different approaches have been proposed to optimise this coupling method, however, in this work we did not attempt to further optimise the coupling via a programming interface. Our work shows clearly that the coupling through TTMs has many advantages compared to one using a programming interface.