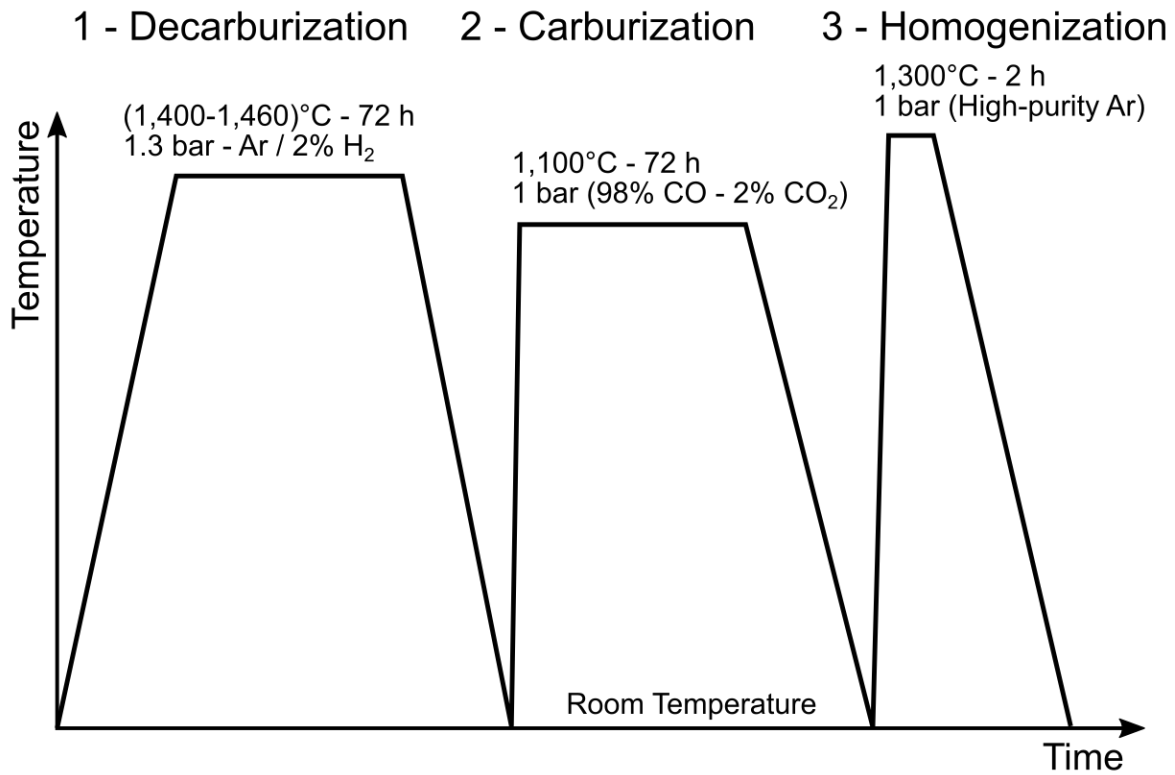


Supplementary Fig. 1.

Schematic illustration of the different fabrication steps of the compositionally graded

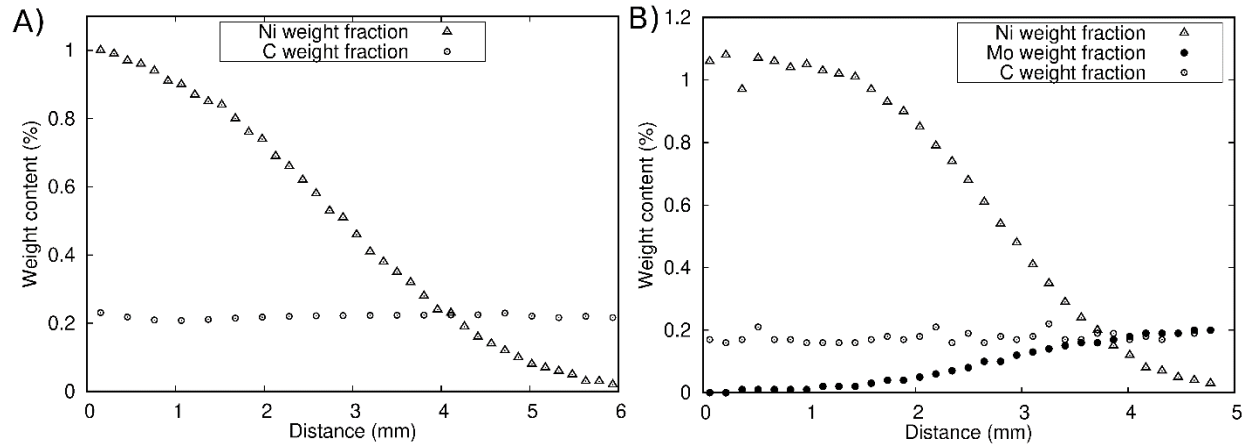
samples. 1- Hot uniaxial compression. 2- High temperature decarburization/diffusion treatment.

3- Re-carburization and homogenization treatments. 4- Grain size refinement.



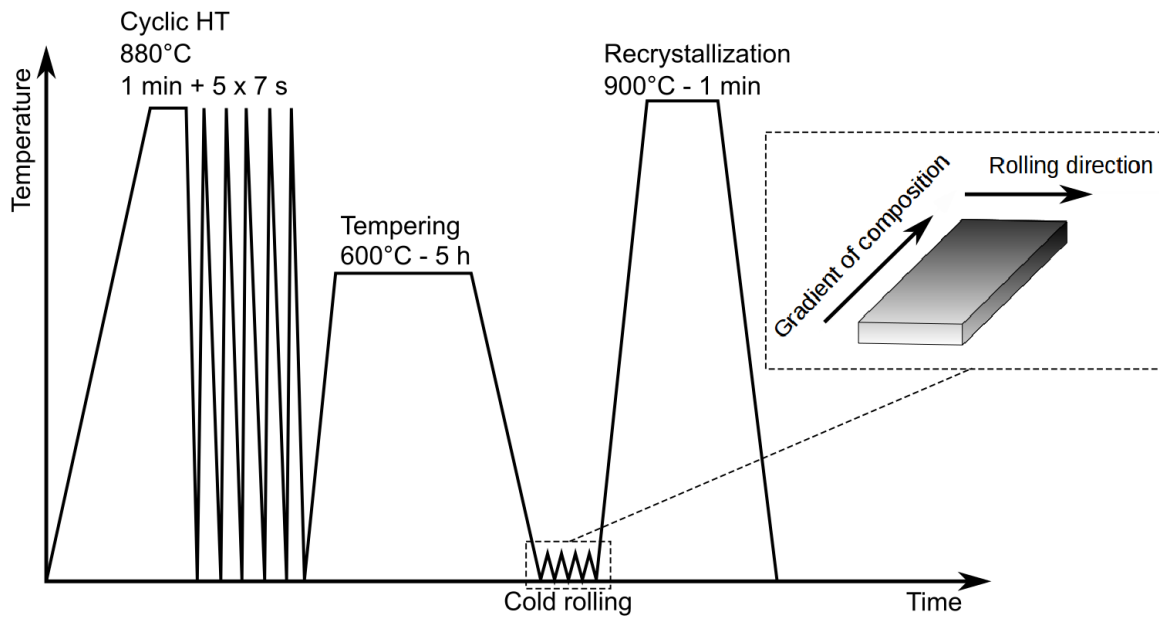
Supplementary Fig. 2.

Thermal schedule across the different heat treatments required for the establishment of the composition gradients. 1- Decarburization/diffusion treatment to generate millimeter scale composition gradients, 2- Re-carburization treatment to introduce the required amount of carbon, and 3- Homogenization of carbon across the sample.



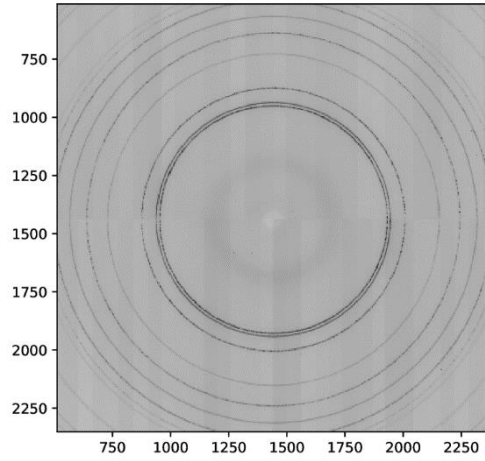
Supplementary Fig. 3.

Composition profiles in diffusion couples, measured using EPMA after decarburization and recarburization treatments. (A) Fe-C/Fe-C-1Ni (wt. %) couple. (B) Fe-C-1Ni/Fe-C-0.2Mo (wt. %) couple.



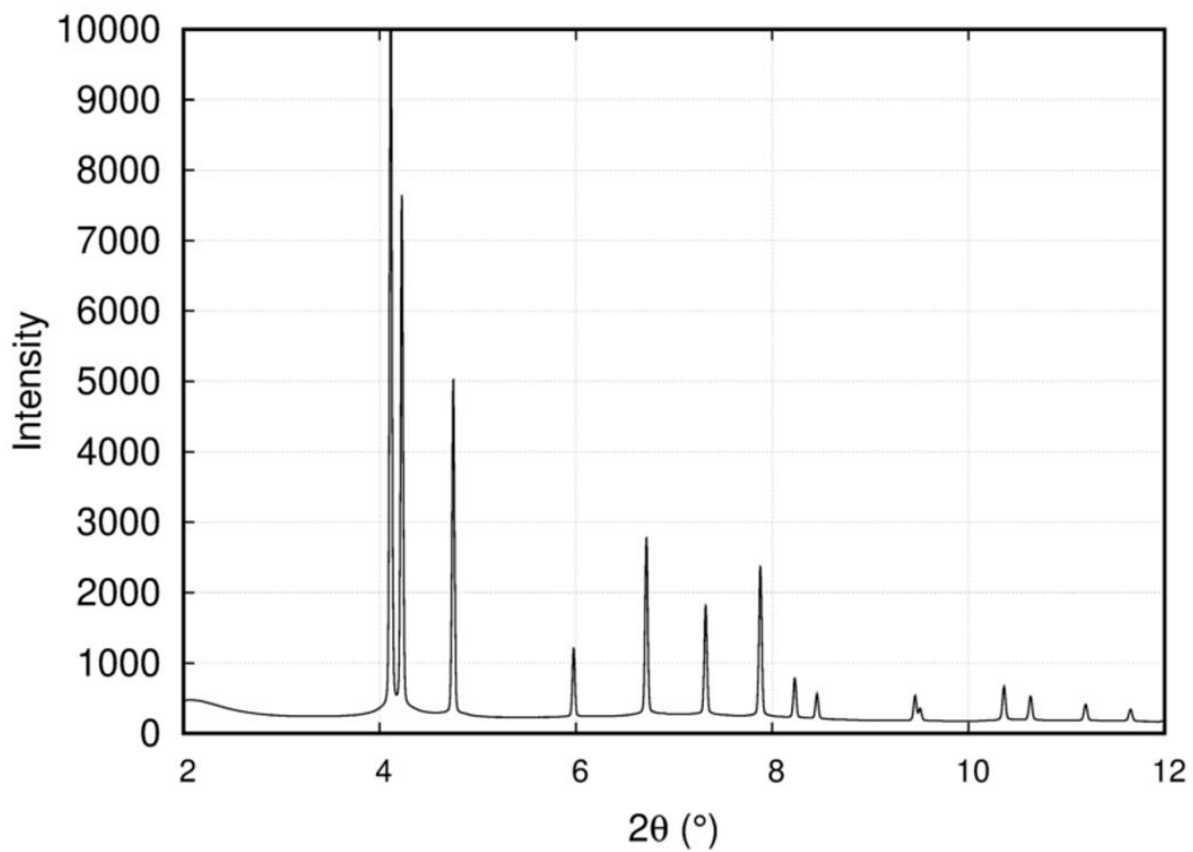
Supplementary Fig. 4.

Schematic illustration of the different steps used for grain size refinement. The thermomechanical schedule includes cyclic heat treatments, tempering, cold rolling and recrystallization. During rolling, the composition gradient in the sample is kept perpendicular to the rolling direction.



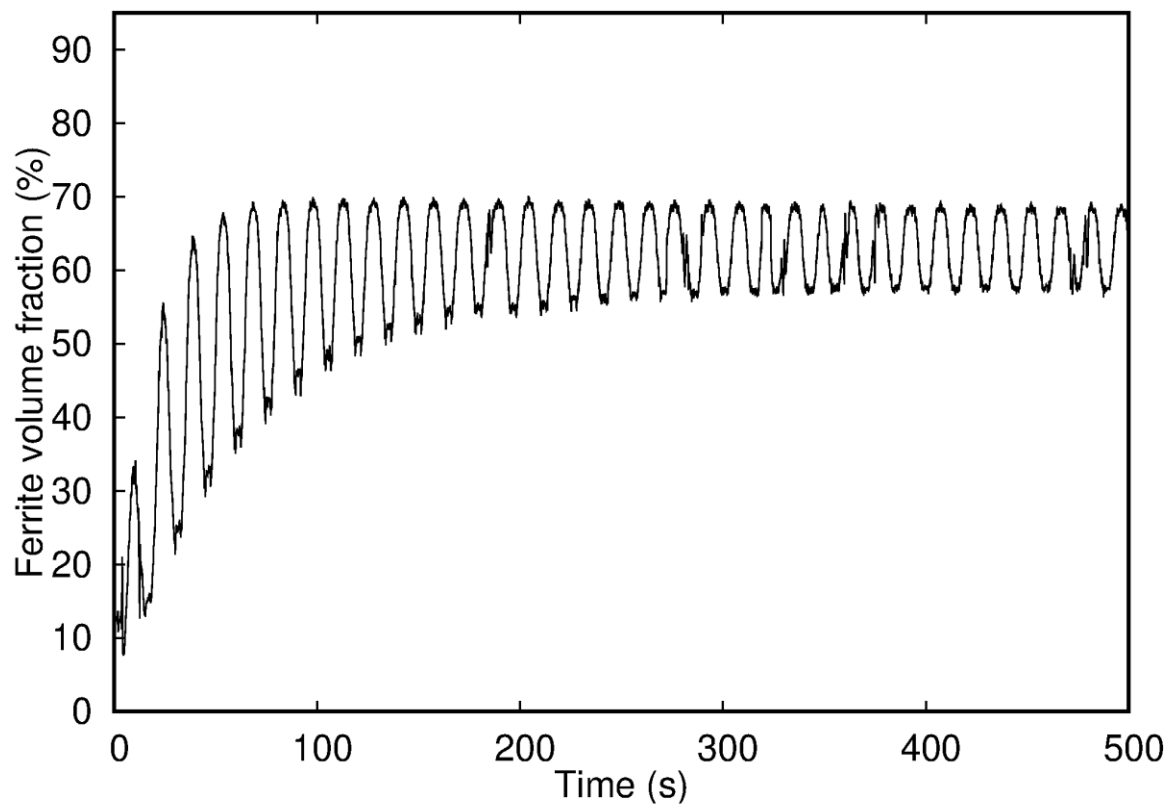
Supplementary Fig. 5.

Example of Debye-Scherrer diffraction rings as recorded by the detector. 1D diffractogram (intensity vs 2θ) resulting from the circular integration of (A).



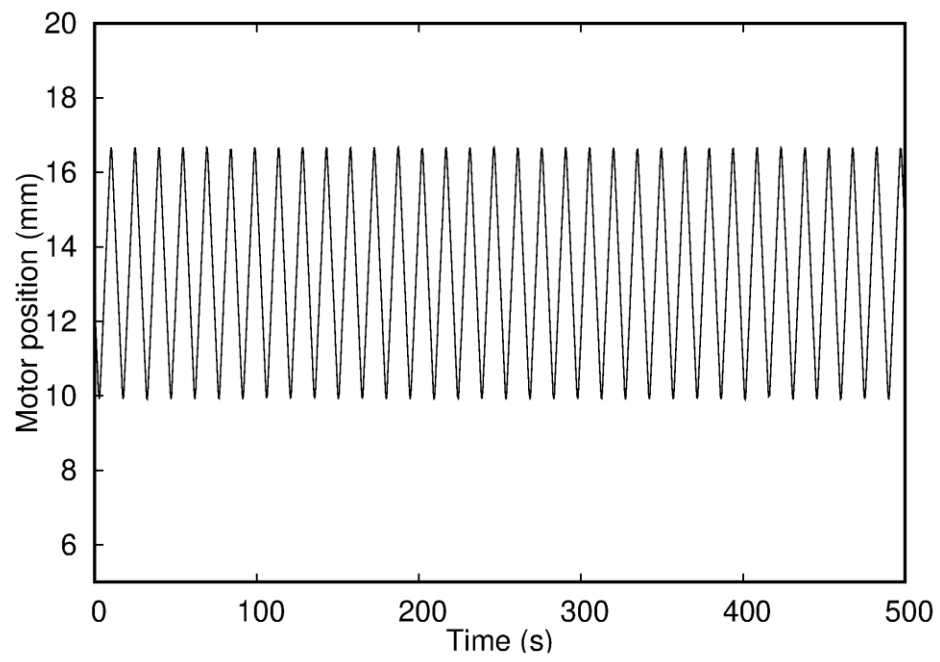
Supplementary Fig. 6.

1D diffractogram (intensity vs 2θ) resulting from the circular integration of Supplementary Fig. 5.



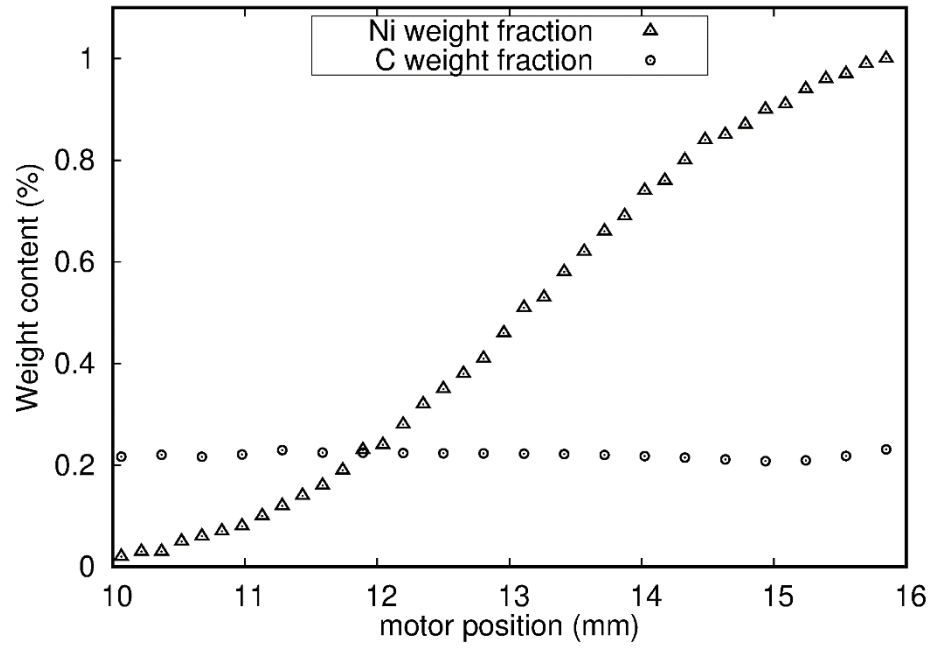
Supplementary Fig. 7.

Evolution of the ferrite fraction as function of time during the *in situ* run of the Fe-C/Fe-C-Ni diffusion couple at 730°C



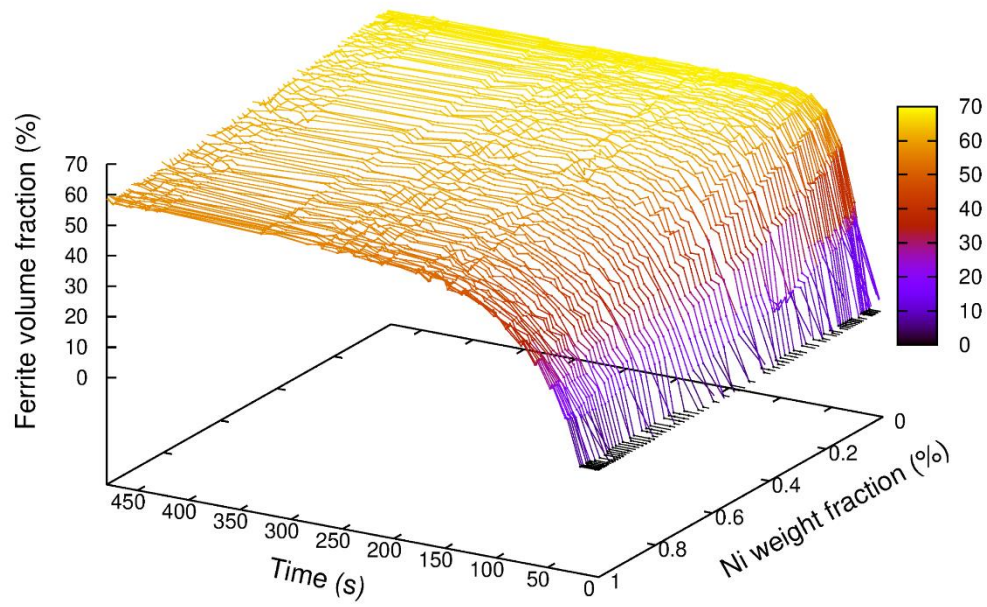
Supplementary Fig. 8.

Recorded motor position as function of time during the HEXRD experiment corresponding to
Supplementary Fig. 7.



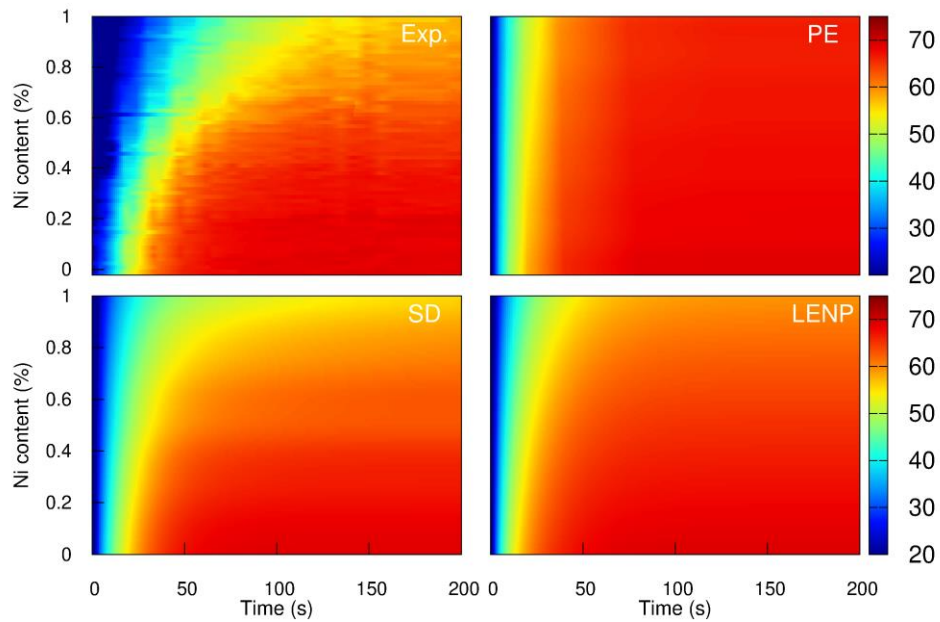
Supplementary Fig. 9.

Nickel and carbon contents as function of position in the sample used during the experiment corresponding to Supplementary Fig. 7.



Supplementary Fig. 10.

Visualization of the final data set corresponding to Supplementary Fig. 7, after the data treatment procedure is complete.



Supplementary Fig. 11.

Heat maps representing the evolution of ferrite fraction as function of time and nickel composition. High-energy X-ray diffraction measurements (Exp.) are presented alongside calculation results from solute drag (SD), paraequilibrium (PE) and local equilibrium negligible partitioning (LENP) models.