

Supplementary Materials

Novel Hybrid Ferromagnetic Fe–Co/Nanodiamond Nanostructures: Influence of Carbon on Their Structural and Magnetic Properties

Panagiotis G. Ziogas ¹, Athanasios B. Bourlino ¹, Polyxeni Chatzopoulou ², George P. Dimitrakopoulos ², Anastasios Markou ¹ and Alexios P. Douvalis ^{1,*}

¹ Physics Department, University of Ioannina, 45110 Ioannina, Greece; p.ziogas@uoi.gr (P.G.Z.); bourlino@uoi.gr (A.B.B.); amarkou@uoi.gr (A.M.); adouval@uoi.gr (A.P.D.)

² Physics Department, Aristotle University of Thessaloniki, 54124 Thessaloniki, Greece; pochatzo@physics.auth.gr (P.C.); gdim@auth.gr (G.P.D.)

* Correspondence: adouval@uoi.gr

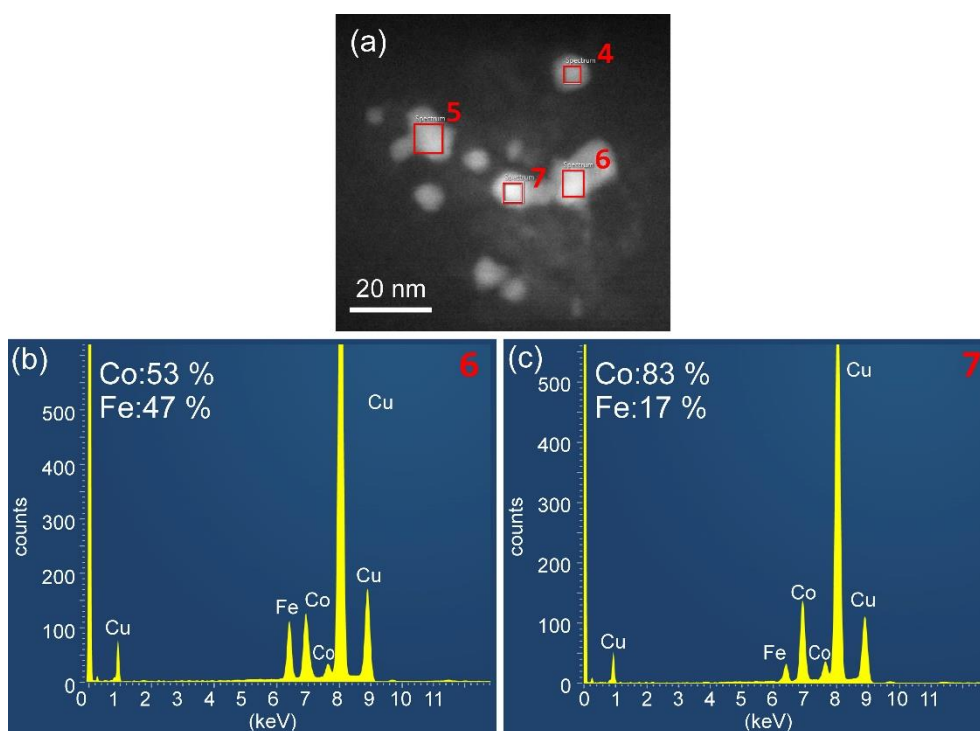


Figure S1. (a) HAADF-STEM image from the NHD-700,30m, illustrating some locations where EDS analysis was conducted. Respective values are listed in Table S1. (b,c) spectra #6 and #7 from NPs with high and low Fe-composition respectively (refer to Table S1 for results).

Table S1. Fe compositions measured in NHD-700,30m, using EDS analysis on individual Fe-Co NPs.

Point	Fe at. %
1	32
2	46
3	38
4	44
5	32
6	47
7	17
8	15
9	36
10	39
11	36
12	41
13	37
Average:	35

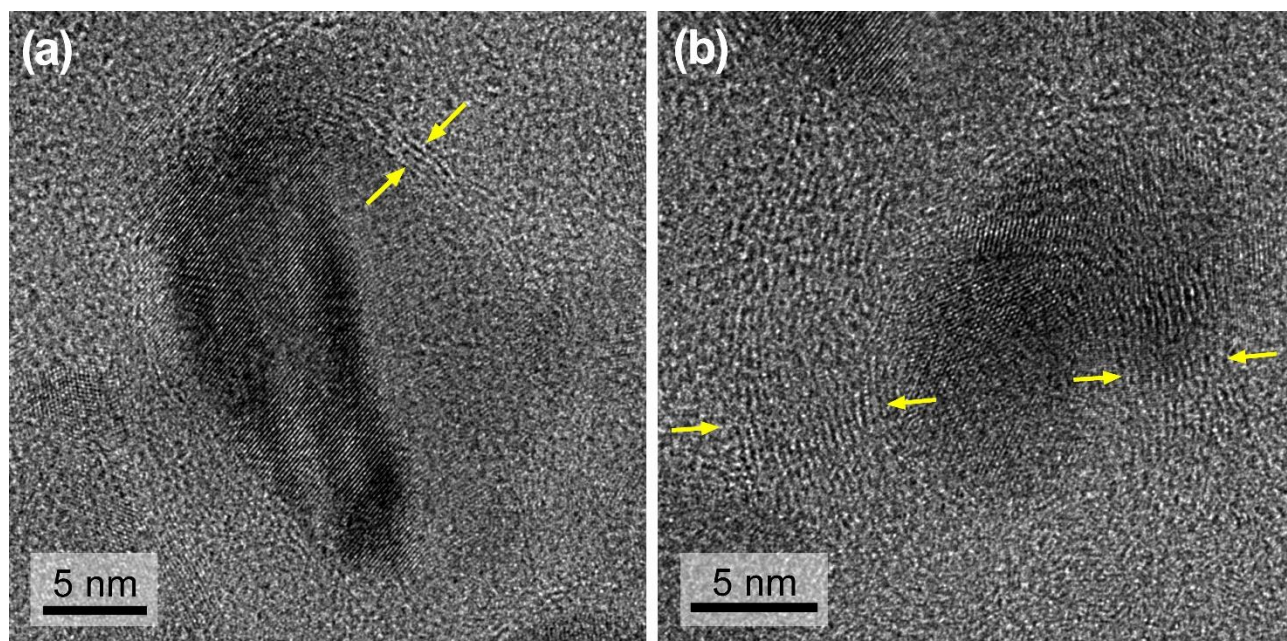


Figure S2. HRTEM images obtained from sample NHD-700,30m, showing graphitic-type layers wrapping around the Fe-Co NPs (yellow arrows).

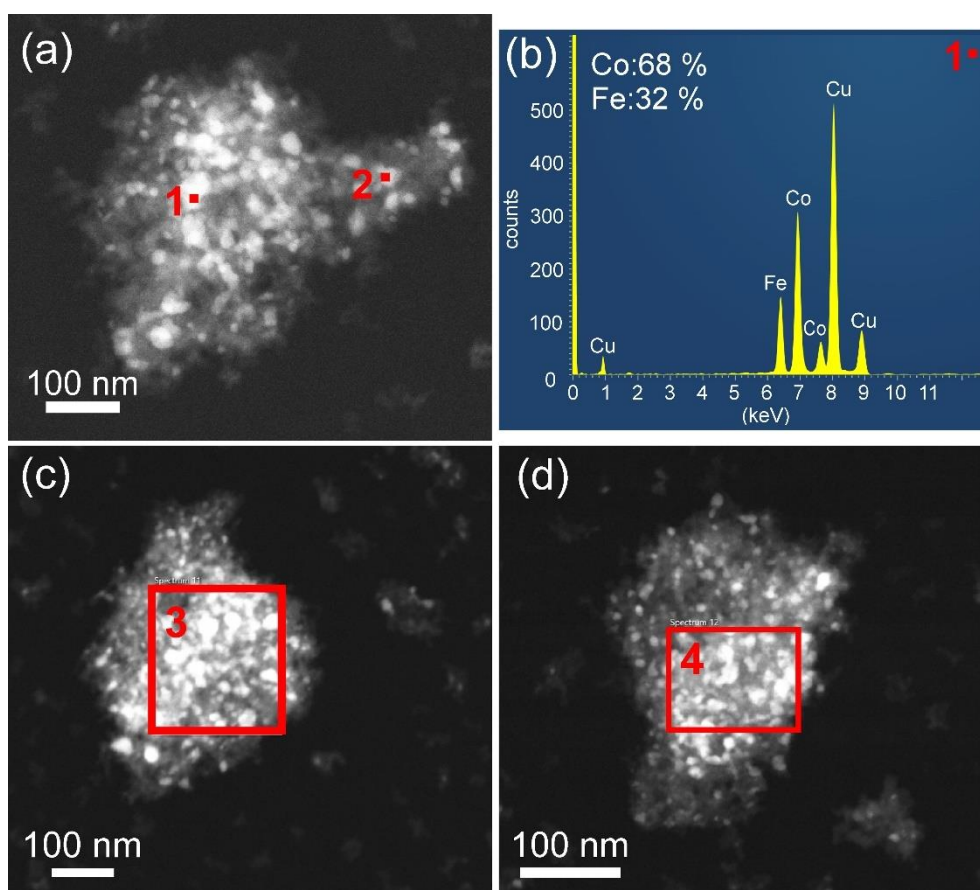


Figure S3. (a) HAADF-STEM image from the En-NHD-700,30m-SC, showing positions where point-EDS spectra were collected. (b) The point spectrum from position 1 and corresponding atomic percentage. (c,d) HAADF-STEM images showing other areas of interest (refer to Table S2 for all results).

Table S2. Fe compositions derived from point and areal EDS spectra obtained from sample En-NHD-700,30m-SC.

Point/Area	Fe at %
point 1	32
point 2	33
area 3	35
area 4	33
Average	33

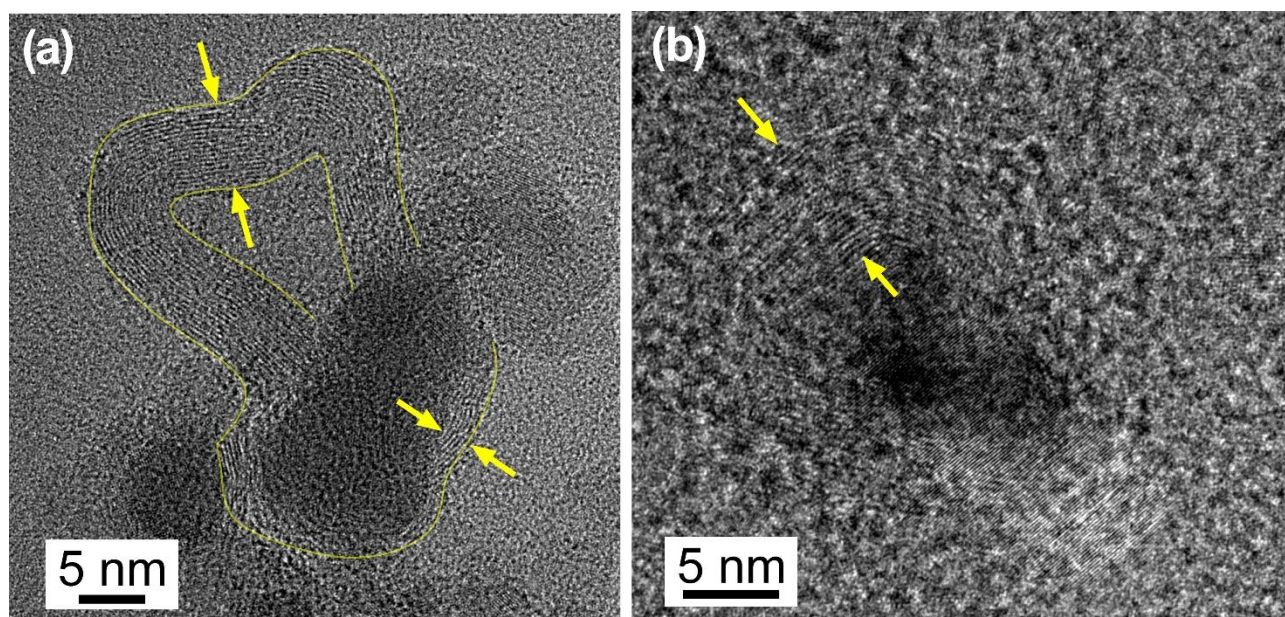


Figure S4. HRTEM images obtained from sample En-NHD-700,30m-SC, showing graphitic-type layers (yellow arrows) wrapping around FeCo NPs. Notably, in (a), these layers are observed to extend partially away from the NP.

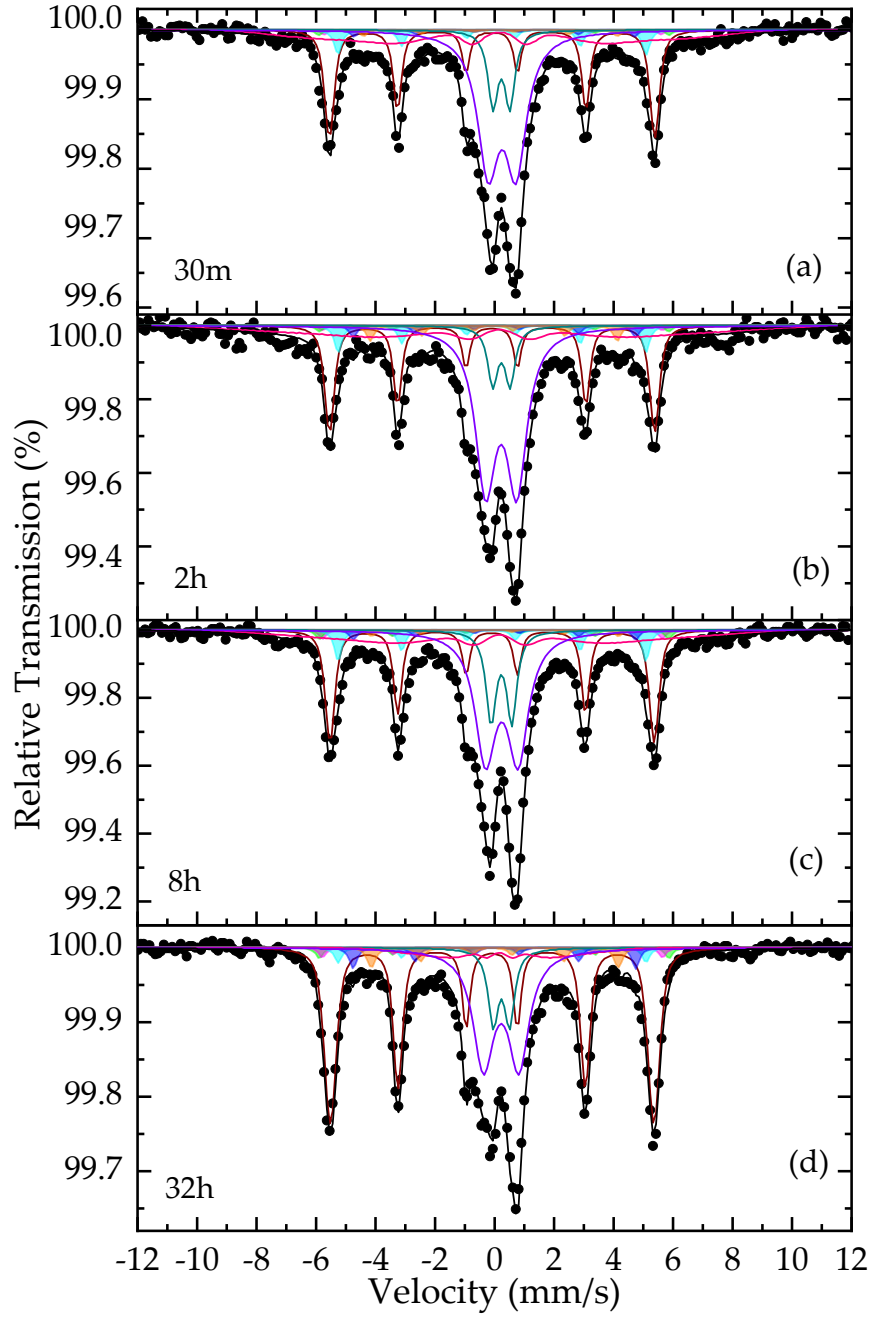


Figure S5. Fitted ^{57}Fe Mössbauer spectra of the NHD-600,30m (a), NHD-600,2h (b), NHD-600,8h (c) and NHD-600,32h (d) samples collected at RT.

Table S3. Mössbauer hyperfine parameters as resulting from the best fits of the corresponding spectra of the samples shown on **Figure S5**. IS is the isomer shift (given relative to $\alpha\text{-Fe}$ at 300 K), $\Gamma/2$ is the half line-width, QS is the quadrupole splitting, 2ϵ is the quadrupole shift, B_{hf}^{C} is the central value of the hyperfine magnetic field, ΔB_{hf} is the total spreading (Gaussian-type) of the B_{hf} values around the central B_{hf}^{C} value, and AA is the relative spectral absorption area of each component used to fit the spectra. Typical errors are ± 0.02 mm/s for IS, $\Gamma/2$, 2ϵ and QS, ± 3 kOe for B_{hf}^{C} and $\pm 3\%$ for AA.

Sample	Component	IS (mm/s)	$\Gamma/2$ (mm/s)	QS or 2ϵ (mm/s)	B_{hf}^{C} (kOe)	ΔB_{hf} (kOe)	Area (%)	Color
NHD-600,30m	$\text{Fe}_{35}\text{Co}_{65}$	0.02	0.15	0.02	340	5	26	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	366	0	1	Green

	Martensitic Fe-Co (2)	0.04	0.15	0.01	352	0	1	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	322	0	5	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	293	0	1	Blue
	Martensitic Fe-Co (5)	0.05	0.15	-0.15	265	0	1	Orange
	SPM Fe ³⁺ (1)	0.34	0.2	0.57	0	0	10	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	355	104	17	Pink
	SPM Fe ³⁺ (2)	0.36	0.427	0.94	0	0	37	Purple
NHD-600,2h	Fe ₃₅ Co ₆₅	0.02	0.15	0.02	340	5	24	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	1	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	1	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	321	0	5	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.01	258	0	3	Orange
	SPM Fe ³⁺ (1)	0.34	0.2	0.58	0	0	7	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	393	158	17	Pink
	SPM Fe ³⁺ (2)	0.33	0.41	1.05	0	0	39	Purple
NHD-600,8h	Fe ₃₅ Co ₆₅	0.02	0.15	0.02	339	5	26	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	321	0	6	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	0	1	Orange
	SPM Fe ³⁺ (1)	0.34	0.2	0.70	0	0	11	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	334	133	17	Pink
	SPM Fe ³⁺ (2)	0.35	0.42	1.09	0	0	33	Purple
NHD-600,32h	Fe ₃₅ Co ₆₅	0.02	0.15	0.02	338	8	43	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	321	0	3	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	4	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	3	4	Orange
	SPM Fe ³⁺ (1)	0.34	0.20	0.55	0	0	9	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	162	60	6	Pink
	SPM Fe ³⁺ (2)	0.33	0.42	1.19	0	0	28	Purple

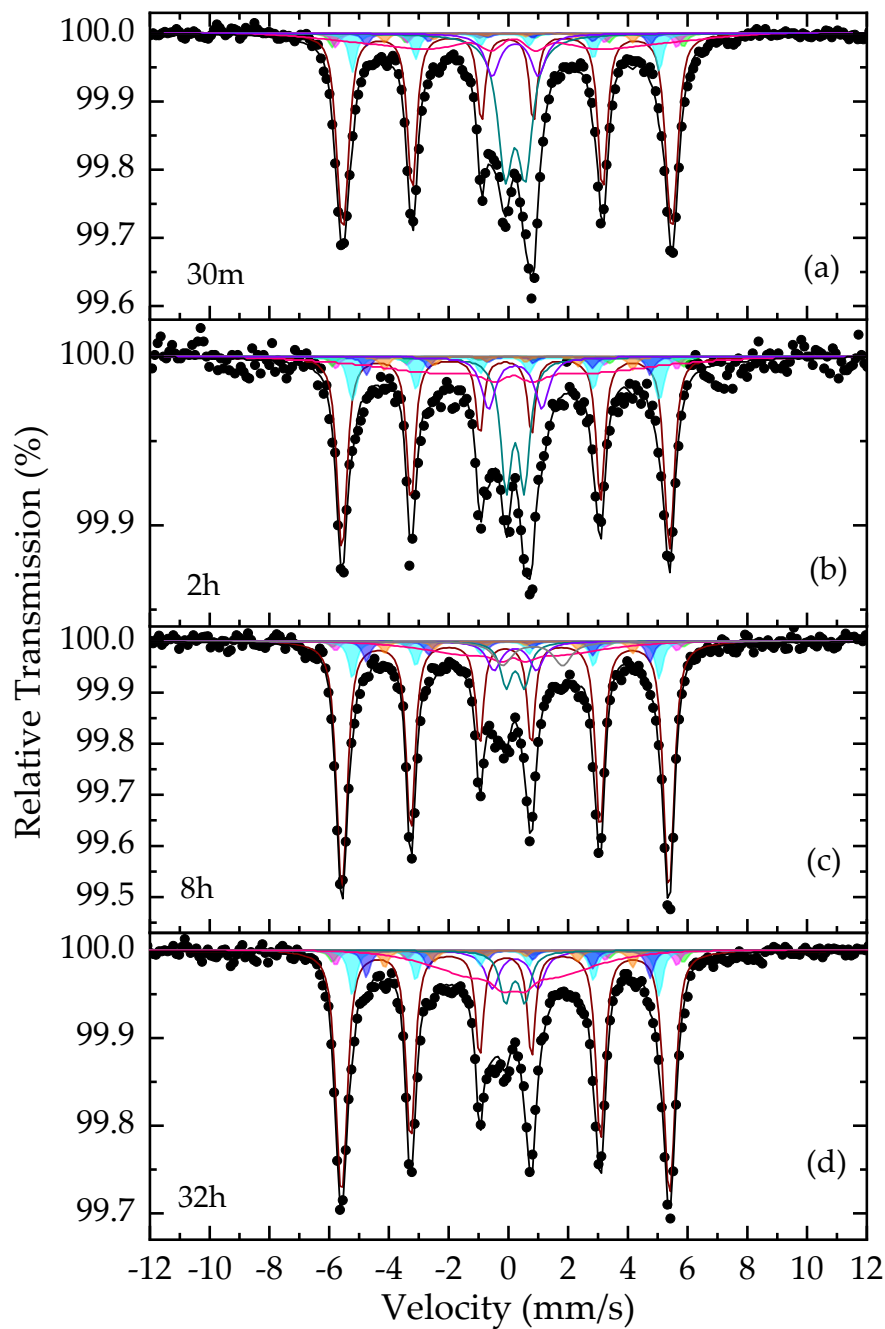


Figure S6. Fitted ^{57}Fe Mössbauer spectra of the NHD-650,30m (a), NHD-650,2h (b), NHD-650,8h (c) and NHD-650,32h (d) samples collected at RT.

Table S4. Mössbauer hyperfine parameters as resulting from the best fits of the corresponding spectra of the samples shown on **Figure S6**. IS is the isomer shift (given relative to $\alpha\text{-Fe}$ at 300 K), $\Gamma/2$ is the half line-width, QS is the quadrupole splitting, 2ϵ is the quadrupole shift, B_{hf}^{C} is the central value of the hyperfine magnetic field, ΔB_{hf} is the total spreading (Gaussian-type) of the B_{hf} values around the central B_{hf}^{C} value, and AA is the relative spectral absorption area of each component used to fit the spectra. Typical errors are ± 0.02 mm/s for IS, $\Gamma/2$, 2ϵ and QS, ± 3 kOe for B_{hf}^{C} and $\pm 3\%$ for AA.

Sample	Component	IS (mm/s)	$\Gamma/2$ (mm/s)	QS or 2ϵ (mm/s)	B_{hf}^{C} (kOe)	ΔB_{hf} (kOe)	Area (%)	Color
NHD-650,30m	$\text{Fe}_{35}\text{Co}_{65}$	0.07	0.15	-0.01	342	8	44	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	2	Green

	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	319	0	6	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	0	2	Orange
	SPM Fe ³⁺ (1)	0.34	0.30	0.67	0	0	21	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	285	105	14	Pink
	SPM Fe ³⁺ (2)	0.34	0.30	1.54	0	0	7	Purple
NHD-650,2h	Fe ₃₅ Co ₆₅	0.02	0.15	0.00	341	5	40	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	319	3	9	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	3	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	0	2	Orange
	SPM Fe ³⁺ (1)	0.34	0.22	0.60	0	0	15	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	218	160	18	Pink
	SPM Fe ³⁺ (2)	0.34	0.28	1.79	0	0	8	Purple
NHD-650,8h	Fe ₃₅ Co ₆₅	0.00	0.15	0.00	340	6	58	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	1	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	319	0	7	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	4	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	0	2	Orange
	SPM Fe ³⁺ (1)	0.34	0.22	0.61	0	0	6	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	134	105	11	Pink
	SPM Fe ³⁺ (2)	0.34	0.28	1.42	0	0	5	Purple
	SPM Fe ²⁺	0.92	0.36	2.00	0	0	5	Grey
NHD-650,32h	Fe ₃₅ Co ₆₅	0.01	0.15	0.00	341	7	53	Maroon
	Martensitic Fe-Co (1)	0.09	0.15	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.15	0.01	355	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.15	0.04	319	0	7	Cyan
	Martensitic Fe-Co (4)	0.15	0.15	-0.08	295	0	4	Blue
	Martensitic Fe-Co (5)	0.08	0.15	0.09	258	0	3	Orange
	SPM Fe ³⁺ (1)	0.34	0.22	0.66	0	0	6	Dark Cyan
	MCOL Fe ³⁺	0.28	0.15	0.00	115	100	17	Pink
	SPM Fe ³⁺ (2)	0.34	0.28	1.53	0	0	6	Purple

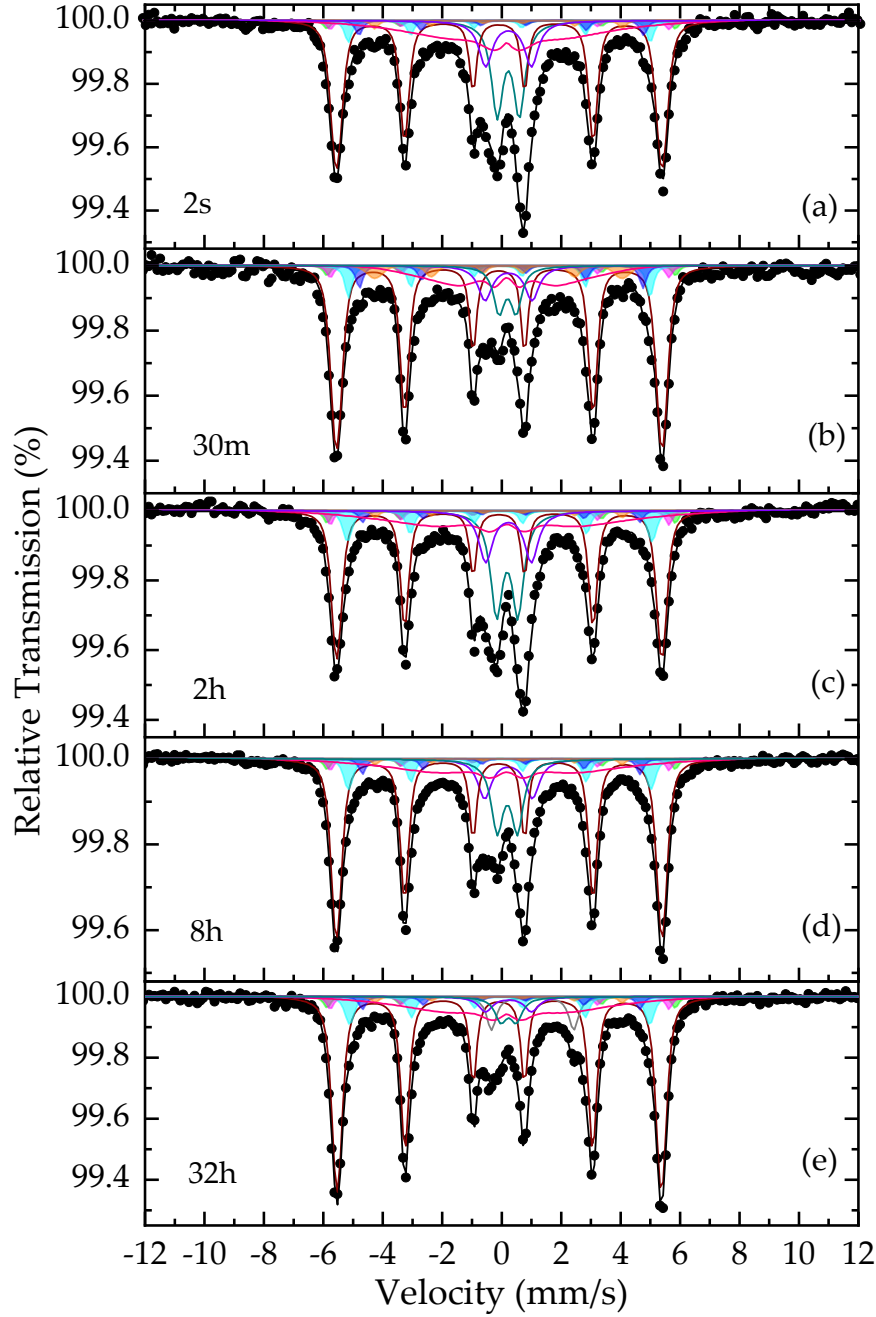


Figure S7. Fitted ^{57}Fe Mössbauer spectra of the NHD-700,2s (a), NHD-700,30m (b), NHD-700,2h (c), NHD-700,8h (d) and NHD-700,32h (e) samples collected at RT.

Table S5. Mössbauer hyperfine parameters as resulting from the best fits of the corresponding spectra of the samples shown on **Figure S7**. IS is the isomer shift (given relative to $\alpha\text{-Fe}$ at 300 K), $\Gamma/2$ is the half line-width, QS is the quadrupole splitting, 2ϵ is the quadrupole shift, B_{hf}^{C} is the central value of the hyperfine magnetic field, ΔB_{hf} is the total spreading (Gaussian-type) of the B_{hf} values around the central B_{hf}^{C} value, and AA is the relative spectral absorption area of each component used to fit the spectra. Typical errors are ± 0.02 mm/s for IS, $\Gamma/2$, 2ϵ and QS, ± 3 kOe for B_{hf}^{C} and $\pm 3\%$ for AA.

Sample	Component	IS (mm/s)	$\Gamma/2$ (mm/s)	QS or 2ϵ (mm/s)	B_{hf}^{C} (kOe)	ΔB_{hf} (kOe)	Area (%)	Color
NHD-700,2s	$\text{Fe}_{35}\text{Co}_{65}$	0.02	0.14	0.02	340	8	44	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	364	0	2	Green

	Martensitic Fe-Co (2)	0.04	0.14	0.01	354	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.14	0.04	316	0	4	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.08	298	0	3	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	259	0	2	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	142	137	20	Pink
	SPM Fe ³⁺ (1)	0.33	0.23	0.75	0	0	15	Dark Cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.54	0	0	9	Purple
NHD-700,30m	Fe ₃₅ Co ₆₅	0.02	0.14	0.02	340	8	52	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	364	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	354	0	2	Magenta
	Martensitic Fe-Co (3)	0.00	0.14	0.04	315	0	6	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.08	296	0	5	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	259	13	5	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	155	65	14	Pink
	SPM Fe ³⁺ (1)	0.30	0.22	0.58	0	0	7	D. cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.58	0	0	7	Purple
NHD-700,2h	Fe ₃₅ Co ₆₅	0.02	0.14	0.02	340	6	39	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	364	0	3	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	354	0	3	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	318	5	7	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.08	290	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	258	0	1	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	212	120	18	Pink
	SPM Fe ³⁺ (1)	0.29	0.24	0.72	0	0	17	Dark Cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.53	0	0	10	Purple
NHD-700,8h	Fe ₃₅ Co ₆₅	0.02	0.14	0.02	340	7	49	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	364	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	354	0	2	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	316	5	8	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.08	290	0	3	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	258	0	1	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	201	120	16	Pink
	SPM Fe ³⁺ (1)	0.29	0.24	0.68	0	0	12	Dark Cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.61	0	0	8	Purple
NHD-700,32h	Fe ₃₅ Co ₆₅	0.02	0.14	0.02	339	7	55	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	364	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	354	0	3	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	314	5	7	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.08	290	0	3	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	258	0	1	Orange

MCOL Fe ³⁺	0.28	0.14	0.00	165	120	17	Pink
SPM Fe ³⁺ (1)	0.34	0.24	0.52	0	0	4	Dark Cyan
SPM Fe ³⁺ (2)	0.34	0.28	1.55	0	0	3	Purple
SPM Fe ²⁺	1.14	0.19	2.76	0	0	5	Grey

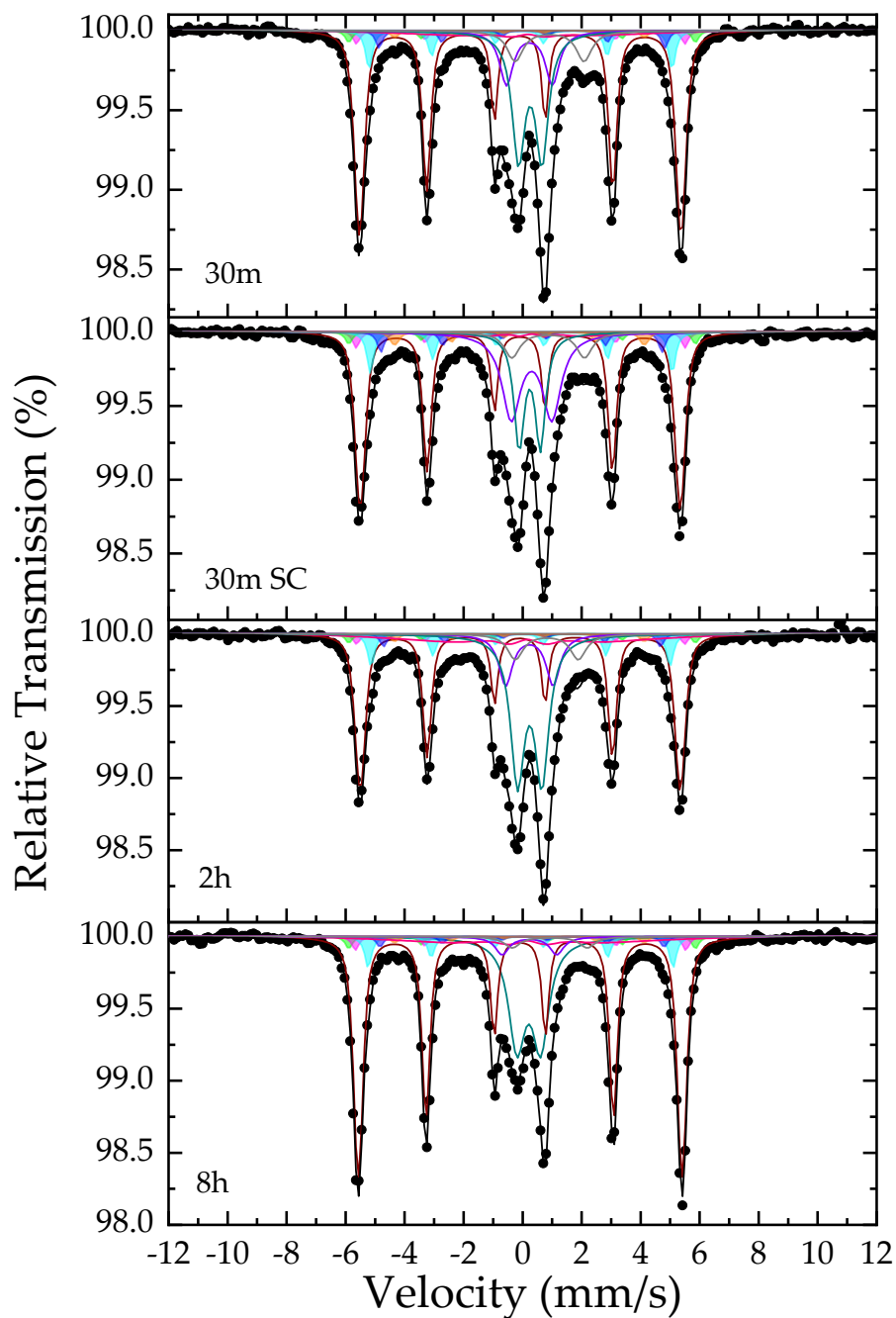


Figure S8. Fitted ⁵⁷Fe Mössbauer spectra of the ⁵⁷Fe enriched En-NHD-700,30m (a), En-NHD-700,30m-SC (b), En-NHD-700,2h (c) and En-NHD-700,8h (d) samples collected at RT.

Table S6. Mössbauer hyperfine parameters as resulting from the best fits of the corresponding spectra of the samples shown on **Figure S8**. IS is the isomer shift (given relative to α -Fe at 300 K), $\Gamma/2$ is the half line-width, QS is the quadrupole splitting, 2ε is the quadrupole shift, B_{hf}^{C} is the central value of the hyperfine magnetic field, ΔB_{hf} is the total spreading (Gaussian-type) of the B_{hf} values around the central B_{hf}^{C} value, and AA is the relative spectral absorption area of each component used to fit the spectra. Typical errors are ± 0.02 mm/s for IS, $\Gamma/2$, 2ε and QS, ± 3 kOe for B_{hf}^{C} and $\pm 3\%$ for AA.

Sample	Component	IS (mm/s)	$\Gamma/2$ (mm/s)	QS or 2ε (mm/s)	B_{hf}^{C} (kOe)	ΔB_{hf} (kOe)	Area (%)	Color
En-NHD-700,30m	$\text{Fe}_{33}\text{Co}_{67}$	0.02	0.14	0.02	339	6	46	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	346	0	2	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	319	0	7	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.09	301	0	3	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	263	3	1	Orange
	MCOL Fe^{3+}	0.28	0.14	0.00	216	120	5	Pink
	SPM Fe^{3+} (1)	0.36	0.26	0.79	0	0	20	Dark Cyan
	SPM Fe^{3+} (2)	0.34	0.28	1.55	0	0	9	Purple
	SPM Fe^{2+}	1.02	0.32	2.33	0	0	6	Grey
En-NHD-700,30m-SC	$\text{Fe}_{33}\text{Co}_{67}$	0.02	0.14	0.02	337	7	41	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	346	0	3	Magenta
	Martensitic Fe-Co (3)	0.05	0.14	0.04	318	0	7	Cyan
	Martensitic Fe-Co (4)	0.12	0.14	-0.09	296	0	4	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	263	5	3	Orange
	MCOL Fe^{3+}	0.43	0.14	0.00	160	60	2	Pink
	SPM Fe^{3+} (1)	0.35	0.21	0.72	0	0	14	Dark Cyan
	SPM Fe^{3+} (2)	0.41	0.38	1.37	0	0	19	Purple
	SPM Fe^{2+}	0.97	0.32	2.46	0	0	5	Grey
En-NHD-700,2h	$\text{Fe}_{33}\text{Co}_{67}$	0.02	0.14	0.02	337	7	38	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	365	0	2	Green

En-NHD-700,8h	Martensitic Fe-Co (2)	0.04	0.14	0.01	346	0	2	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	315	0	6	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.09	292	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	263	3	2	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	234	120	8	Pink
	SPM Fe ³⁺ (1)	0.35	0.28	0.83	0	0	26	Dark Cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.61	0	0	9	Purple
	SPM Fe ²⁺	0.93	0.32	2.13	0	0	5	Grey
	Fe ₃₃ Co ₆₇	0.02	0.14	0.02	341	5	53	Maroon
	Martensitic Fe-Co (1)	0.09	0.14	0.01	365	0	2	Green
	Martensitic Fe-Co (2)	0.04	0.14	0.01	348	0	2	Magenta
	Martensitic Fe-Co (3)	0.02	0.14	0.04	321	0	6	Cyan
	Martensitic Fe-Co (4)	0.13	0.14	-0.09	299	0	2	Blue
	Martensitic Fe-Co (5)	0.08	0.14	-0.15	263	3	1	Orange
	MCOL Fe ³⁺	0.28	0.14	0.00	234	120	6	Pink
	SPM Fe ³⁺ (1)	0.31	0.34	0.80	0	0	22	Dark Cyan
	SPM Fe ³⁺ (2)	0.34	0.28	1.85	0	0	3	Purple
	SPM Fe ²⁺	1.02	0.32	2.55	0	0	2	Grey

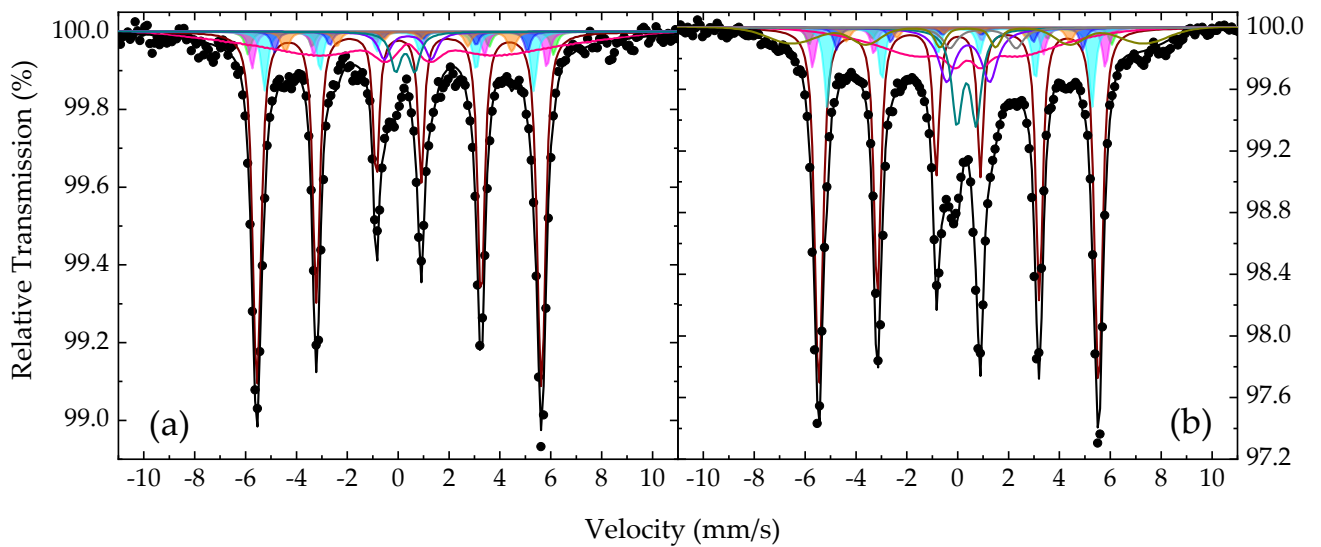


Figure S9. Fitted ⁵⁷Fe Mössbauer spectra of NHD-700,30m sample (a) and of En-NHD-700,30m-SC (b) collected at 11 K.

Table S7. Mössbauer hyperfine parameters as resulting from the best fits of the corresponding spectra of the samples shown on **Figure S9**. IS is the isomer shift (given relative to α -Fe at 300 K), $\Gamma/2$ is the half line-width, QS is the quadrupole splitting, 2ϵ is the quadrupole shift, B_{hf}^C is the central value of the hyperfine magnetic field, ΔB_{hf} is the total spreading (Gaussian-type) of the B_{hf} values around the central B_{hf}^C value, and AA is the relative spectral absorption area of each component used to fit the spectra. Typical errors are ± 0.02 mm/s for IS, $\Gamma/2$, 2ϵ and QS, ± 3 kOe for B_{hf}^C and $\pm 3\%$ for AA.

Sample	Component	IS (mm/s)	$\Gamma/2$ (mm/s)	QS or 2ϵ (mm/s)	B_{hf}^C (kOe)	ΔB_{hf} (kOe)	Area (%)	Color
Non-enriched NHDs- 700,30m	$Fe_{35}Co_{65}$	0.14	0.14	0.02	347	6	52	Maroon
	Martensitic FeCo (1)	0.21	0.14	0.01	373	0	3	Green
	Martensitic FeCo (2)	0.16	0.14	0.01	360	0	4	Magenta
	Martensitic FeCo (3)	0.13	0.14	0.04	329	0	7	Cyan
	Martensitic FeCo (4)	0.26	0.14	-0.08	309	0	2	Blue
	Martensitic FeCo (5)	0.20	0.14	-0.15	277	5	3	Orange
	MCOL Fe^{3+}	0.49	0.14	0.00	323	156	21	Pink
	SPM Fe^{3+} (1)	0.38	0.24	0.75	0	0	5	Dark Cyan
	SPM Fe^{3+} (2)	0.49	0.28	1.85	0	0	4	Purple
Enriched NHDs- 700,30m with SC	$Fe_{33}Co_{67}$	0.14	0.14	0.02	342	5	43	Maroon
	Martensitic FeCo (1)	0.21	0.14	0.01	370	0	2	Green
	Martensitic FeCo (2)	0.16	0.14	0.01	358	0	4	Magenta
	Martensitic FeCo (3)	0.17	0.14	0.04	324	0	8	Cyan
	Martensitic FeCo (4)	0.24	0.14	-0.09	301	0	2	Blue
	Martensitic FeCo (5)	0.20	0.14	-0.15	273	5	2	Orange
	MCOL Fe^{3+}	0.51	0.14	0.00	180	118	15	Pink
	SPM Fe^{3+} (1)	0.46	0.25	0.76	0	0	8	Dark Cyan
	SPM Fe^{3+} (2)	0.52	0.38	1.68	0	0	7	Purple
	SPM Fe^{2+}	1.11	0.34	2.61	0	0	2	Grey
	MRES Fe^{3+}	0.51	0.14	0.00	429	54	9	Dark Yellow

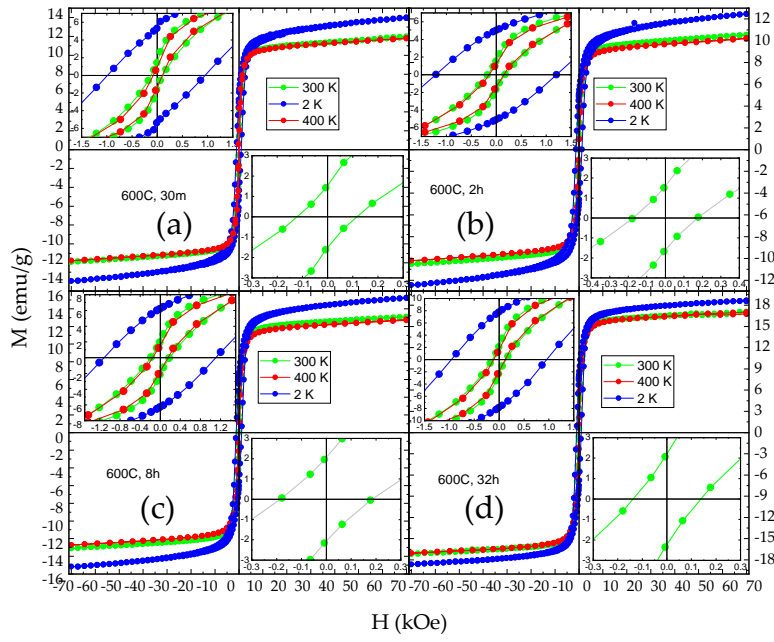


Figure S10. M vs. H isothermal loops of the NHD-600,30m (a), and NHD-600,2h (b), NHD-600,8h (c) and NHD-600,32h (d) samples measured at different temperatures indicated by different colors. The insets in each set of measurements show the details of the loops' characteristics around zero H field for all temperatures (upper left) and with even more detail for the 300 K loop (lower right).

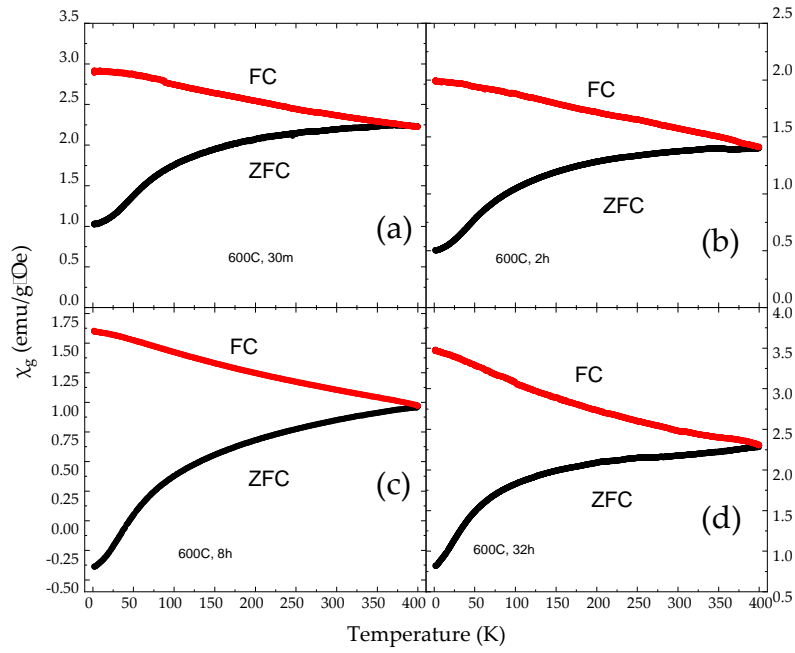


Figure S11. χ_g vs. T measurements of the NHD-600,30m (a), NHD-600,2h (b), NHD-600,8h (c) and NHD-600,32h (d) samples under an applied field of 99 Oe.

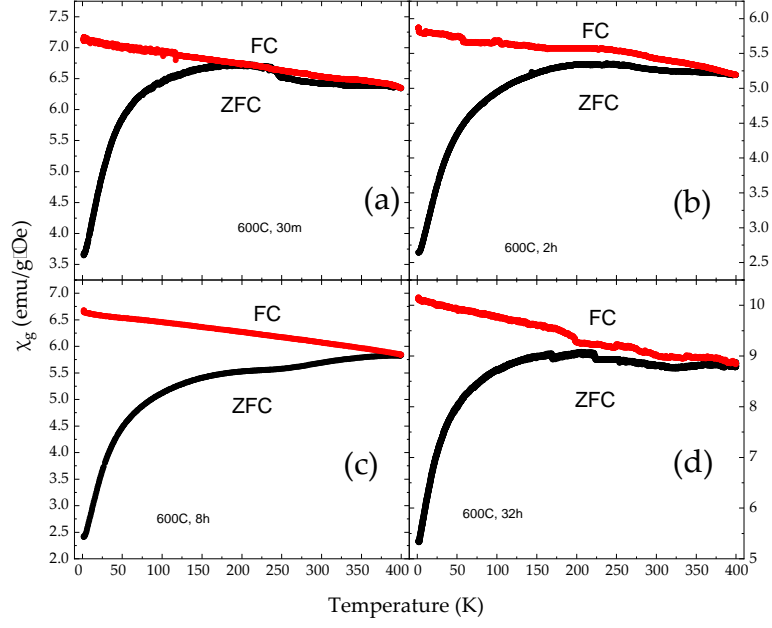


Figure S12. χ_g vs. T measurements of the NHD-600,30m (a), NHD-600,2h (b), NHD-600,8h (c) and NHD-600,32h (d) samples under an applied field of 999 Oe.

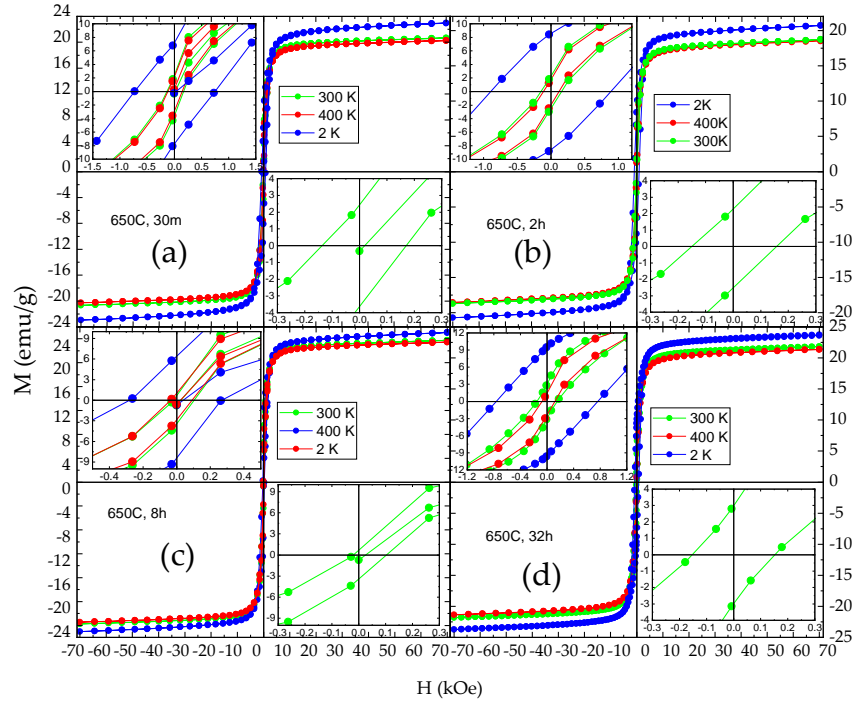


Figure S13. M vs. H isothermal loops of the NHD-650,30m (a), and NHD-650,2h (b), NHD-650,8h (c) and NHD-650,32h (d) samples measured at different temperatures indicated by different colors. The insets in each set of measurements show the details of the loops' characteristics around zero H field for all temperatures (upper left) and with even more detail for the 300 K loop (lower right).

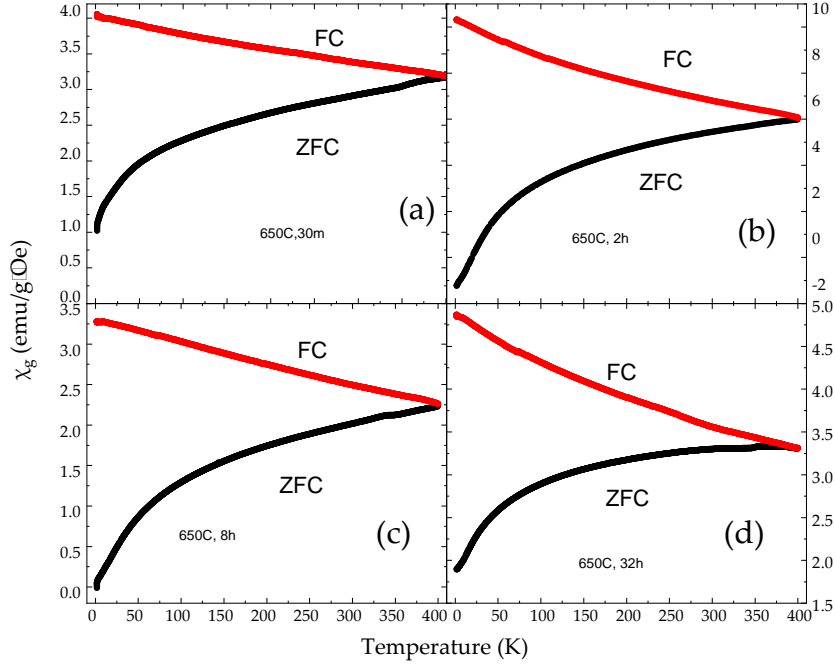


Figure S14. χ_g vs. T measurements of the NHD-650,30m (a), NHD-650,2h (b), NHD-650,8h (c) and NHD-650,32h (d) samples under an applied field of 99 Oe.

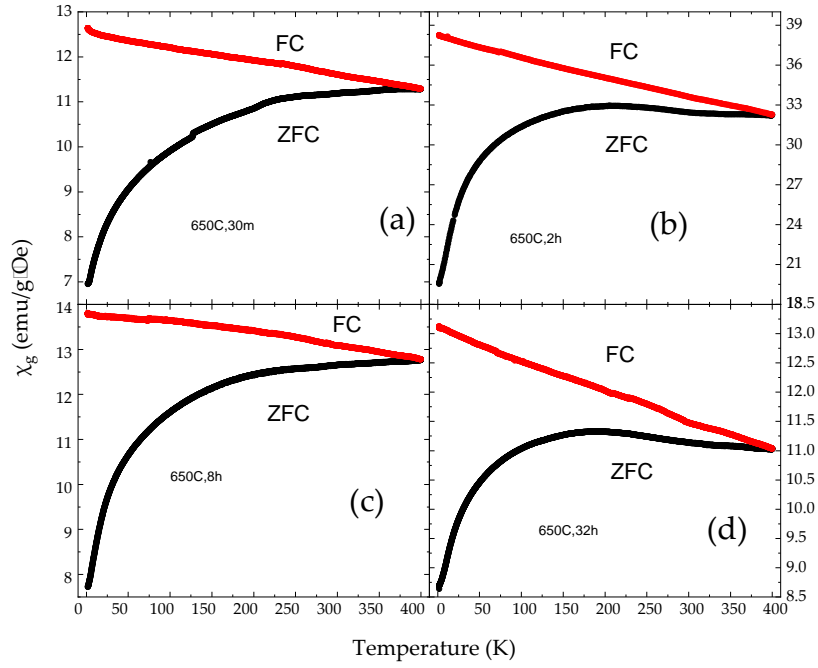


Figure S15. χ_g vs. T measurements of the NHD-650,30m (a), NHD-650,2h (b), NHD-650,8h (c) and NHD-650,32h (d) samples under an applied field of 999 Oe.

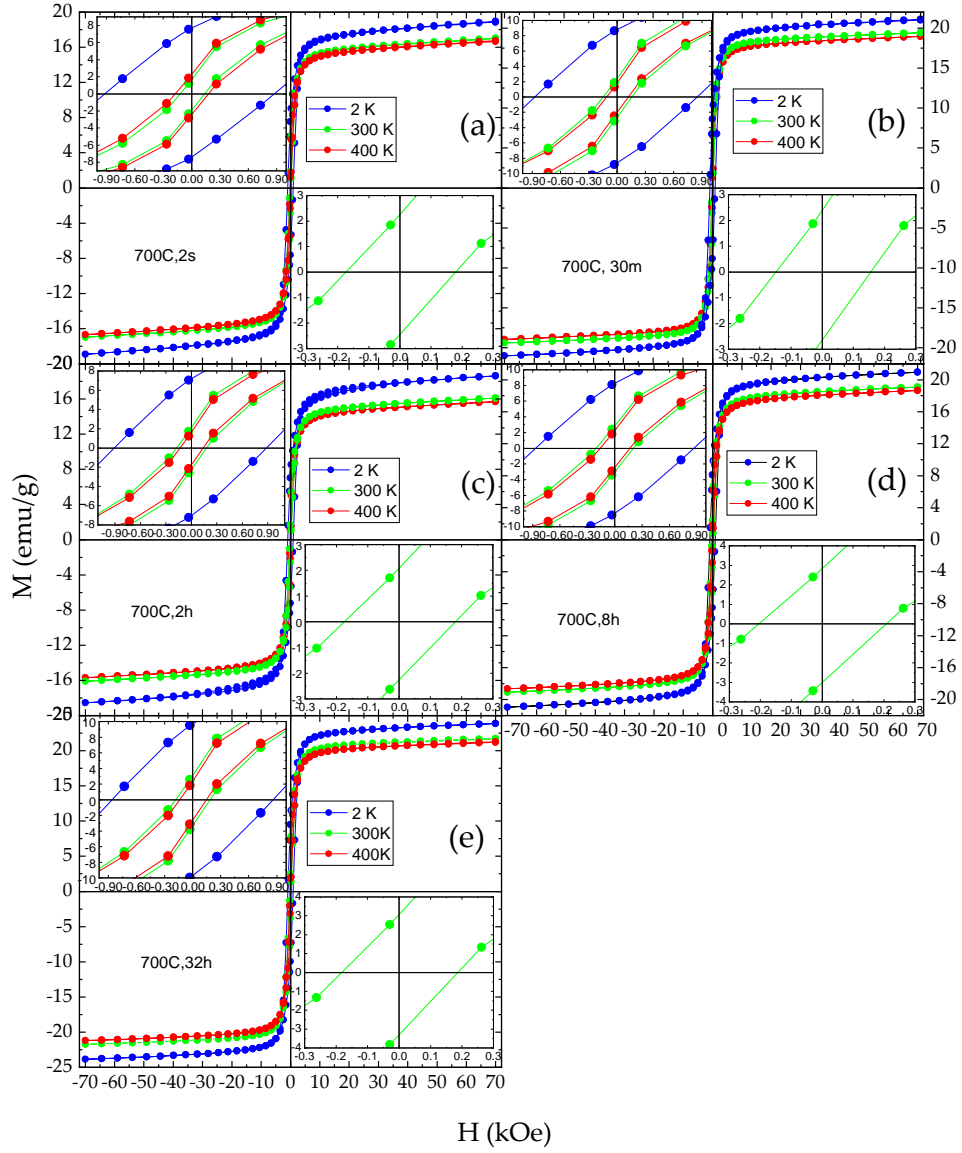


Figure S16. M vs. H isothermal loops of the NHD-700,2s (a), and NHD-700,30m (b), NHD-700,2h (c), NHD-700,8h (d) and NHD-700,32h (e) samples measured at different temperatures indicated by different colors. The insets in each set of measurements show the details of the loops' characteristics around zero H field for all temperatures (upper left) and with even more detail for the 300 K loop (lower right).

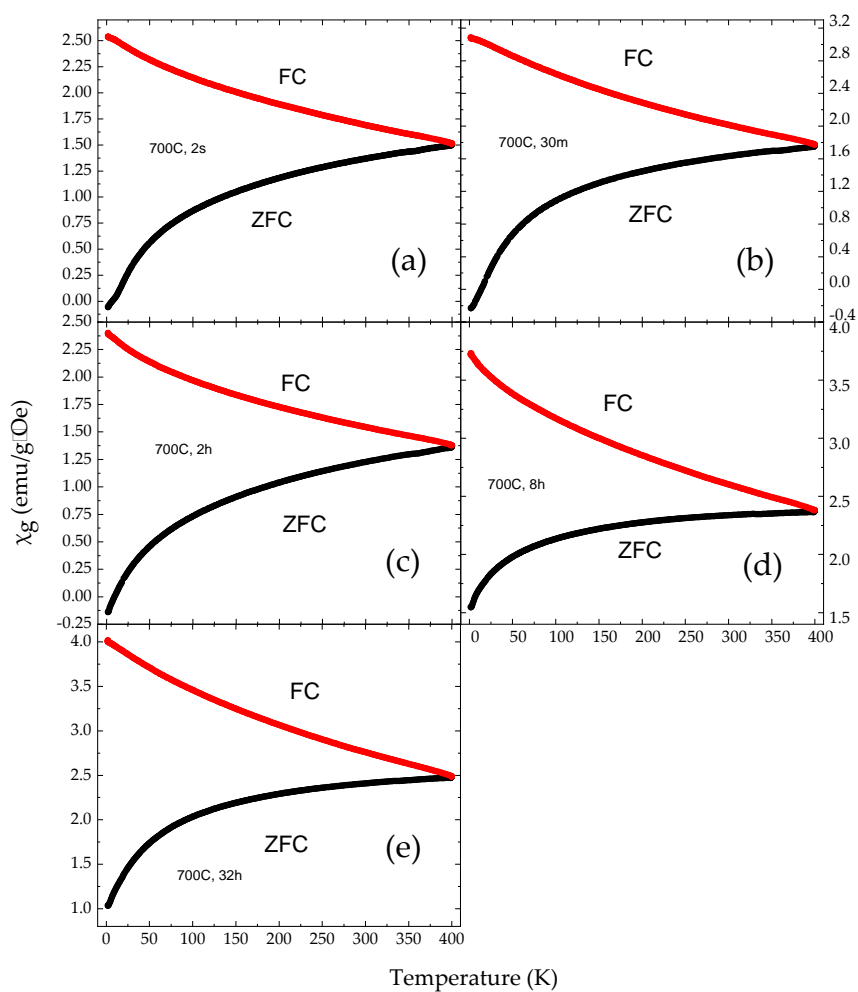


Figure S17. χ_g vs. T measurements of the NHD-700,2s (a), and NHD-700,30m (b), NHD-700,2h (c), NHD-700,8h (d) and NHD-700,32h (e) samples under an applied field of 99 Oe.

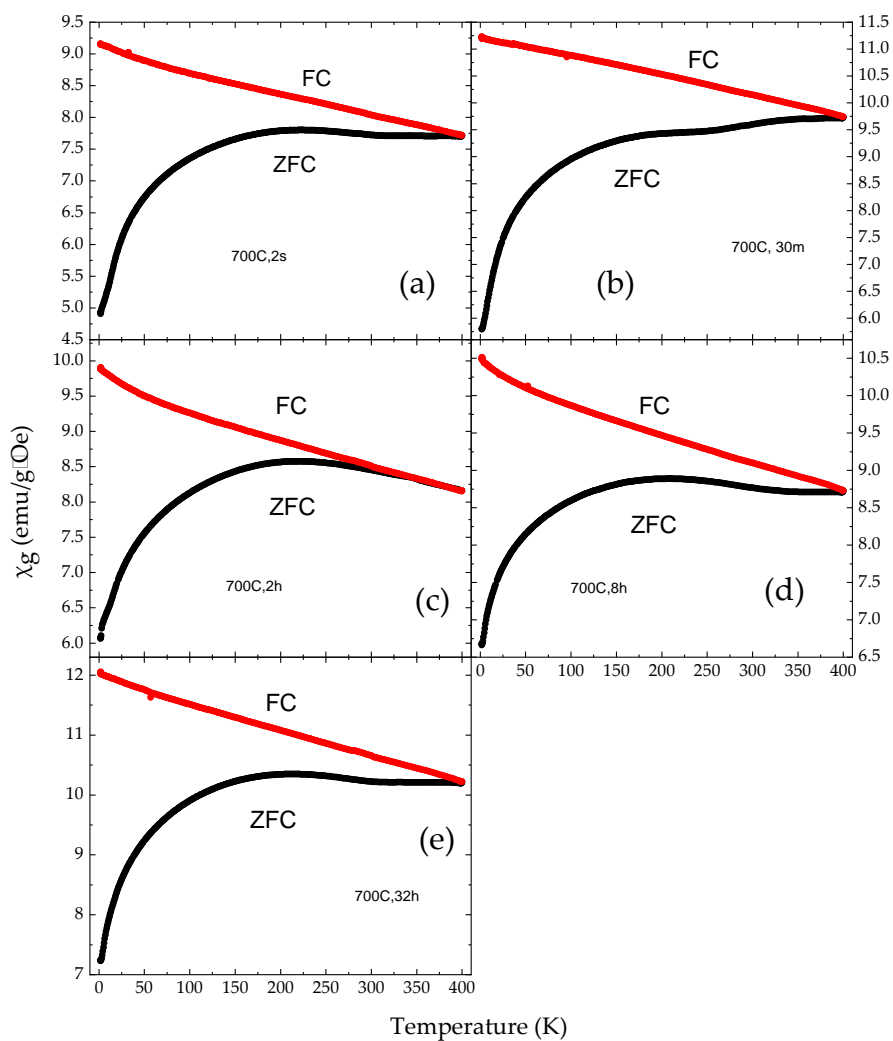


Figure S18. χ_g vs. T measurements of the NHD-700,2s (a), and NHD-700,30m (b), NHD-700,2h (c), NHD-700,8h (d) and NHD-700,32h (e) samples under an applied field of 999 Oe.

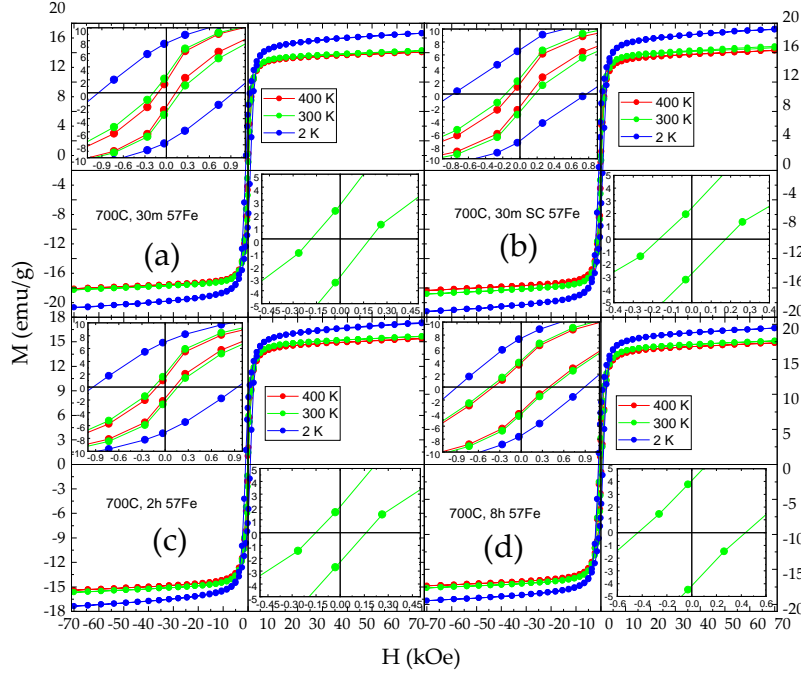


Figure S19. M vs. H isothermal loops of the ^{57}Fe enriched En-NHD-700,30m (a), and En-NHD-700,30m-SC (b), En-NHD-700,2h (c) and En-NHD-700,8h (d) samples measured at different temperatures indicated by different colors. The insets in each set of measurements show the details of the loops' characteristics around zero H field for all temperatures (upper left) and with even more detail for the 300 K loop (lower right).

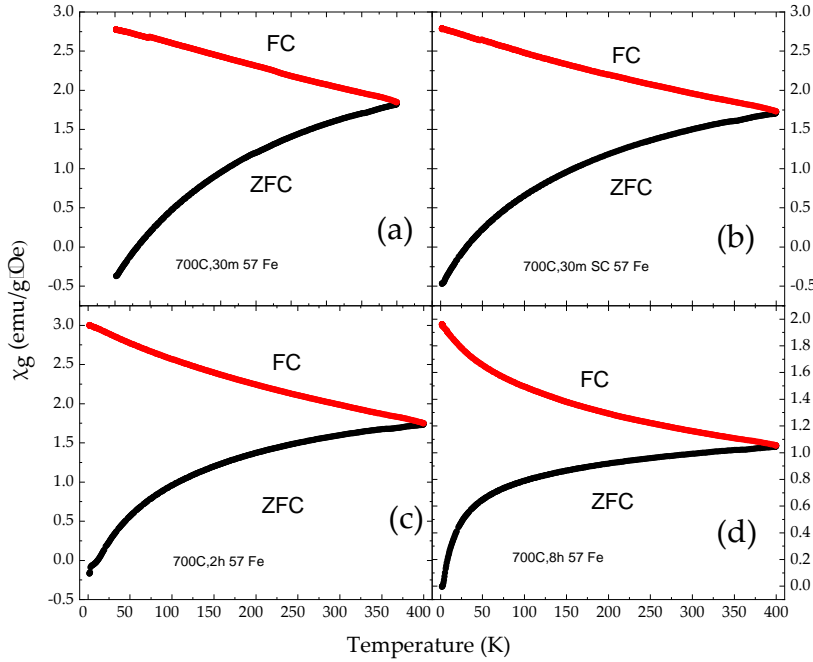


Figure S20. χ_g vs. T measurements of the ^{57}Fe enriched En-NHD-700,30m (a), and En-NHD-700,30m-SC (b), En-NHD-700,2h (c) and En-NHD-700,8h (d) samples under an applied field of 99 Oe.

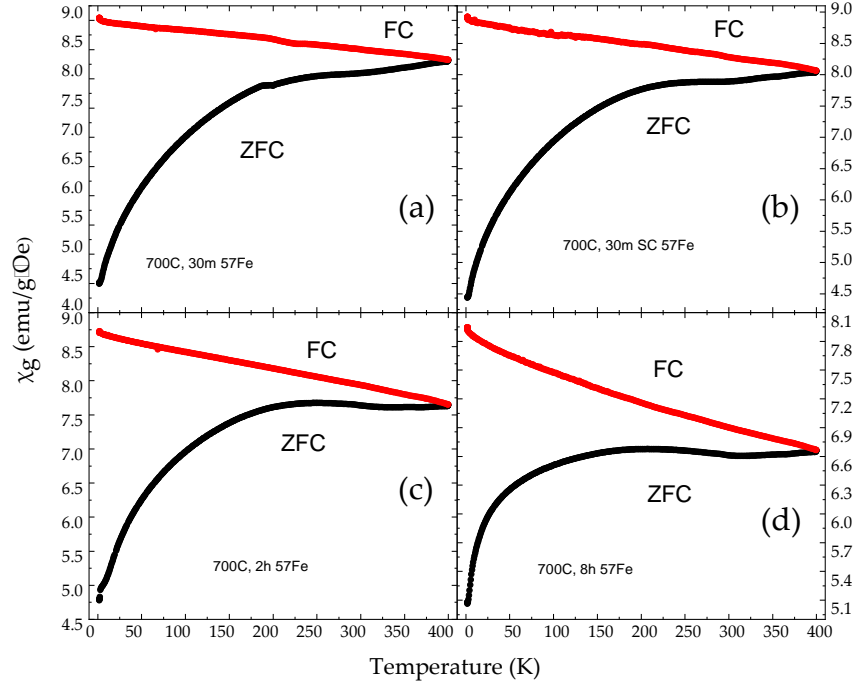


Figure S21. χ_g vs. T measurements of the ^{57}Fe enriched En-NHD-700,30m (a), and En-NHD-700,30m-SC (b), En-NHD-700,2h (c) and En-NHD-700,8h (d) samples under an applied field of 999 Oe.

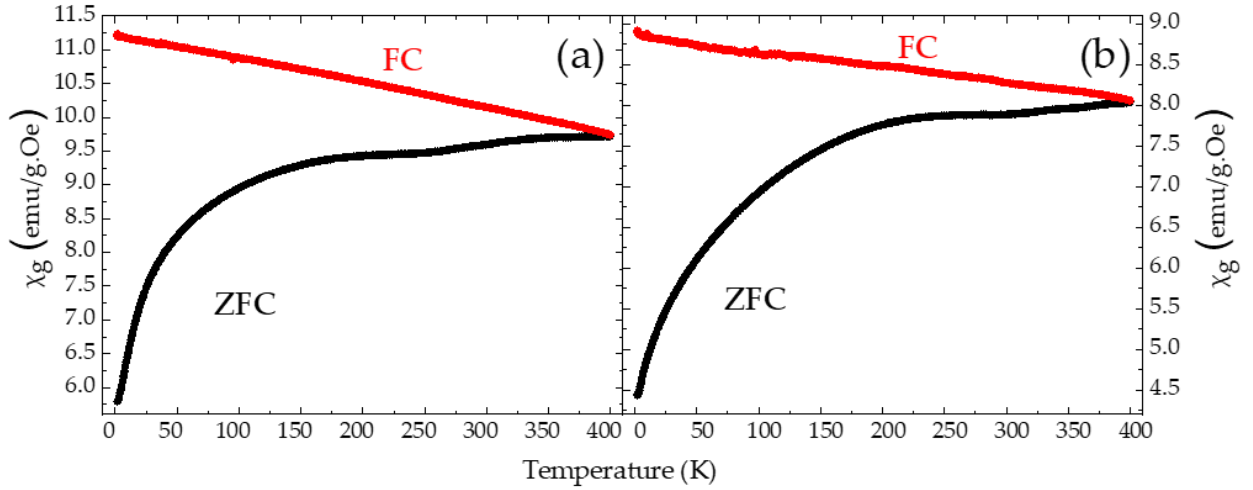


Figure S22. χ_g vs. T measurements of the non-enriched NHD-700,30m (a) and ^{57}Fe enriched En-NHD-700,30m_SC (b) under an applied field of 999 Oe.

Table S8. Magnetic characteristics derived from the respective isothermal loops of all samples at 300 and 2 K.

Sample	T (K)	$M_{\text{max}+}$ (emu/g)	$M_{\text{max}-}$ (emu/g)	$M_{\text{R}+}$ (emu/g)	$M_{\text{R}-}$ (emu/g)	$H_{\text{C}+}$ (Oe)	$H_{\text{C}-}$ (Oe)
NHD-600,30m	300	11.9	-11.9	1.4	-1.6	114	-119
	2	14	-13.9	5.3	-5.3	952	-986
NHD-600,2h	300	10.5	-10.6	1.5	-1.6	183	-168
	2	12.4	-12.4	5	-5.1	1221	-1208
	300	13.1	-13.1	2	-2.1	198	-174

NHD-600,8h	2	15.2	-15.2	6.2	-6.3	1114	-1114
NHD-600,32h	300	17.2	-17.2	2.2	-2.3	140	-130
	2	18.6	-18.7	7.7	-7.8	960	-955
NHD-650,30m	300	20.6	-20.6	2.1	-3.9	167	-138
	2	22.9	-22.9	7	-7.8	749	-729
NHD-650,2h	300	18.7	-18.7	2.2	-2.6	160	-145
	2	20.7	-20.7	8.6	-8.5	895	-895
NHD-650,8h	300	23.8	-23.8	0.8	-3.4	106	-20
	2	25.1	-25.1	6.4	-8.3	265	-271
NHD-650,32h	300	21.8	-21.9	2.8	-3.3	162	-154
	2	23.6	-23.8	9.5	-9.6	768	-756
NHD-700,2s	300	17	-17	1.6	-1.9	140	-120
	2	18.9	-18.9	7.7	-7.4	880	-921
NHD-700,30m	300	19.4	-19.4	2.3	-2.7	160	-152
	2	21.1	-21.1	8.7	-8.6	856	-881
NHD-700,2h	300	16	-16.1	2.1	-2.2	177	-181
	2	18.6	-18.5	7.2	-7.1	892	-903
NHD-700,8h	300	19.1	-19.2	2.8	-3.2	216	-206
	2	20.9	-20.9	8.3	-8.3	872	-869
NHD-700,32h	300	21.7	-21.8	3.1	-3.6	191	-182
	2	23.8	-23.9	9.7	-9.8	866	-857
En-NHD-700,30m	300	16.3	-16.2	2.6	-2.9	205	-180
	2	18.6	-18.6	7.7	-7.6	940	-947
En-NHD-700,30m-SC	300	16.7	-16.8	2.5	-2.7	177	-169
	2	19.2	-19.2	6.8	-7.2	781	-776
En-NHD-700,2h	300	15.7	-15.6	2.1	-2.3	168	-148
	2	17.3	-17.4	7.1	-7	945	-932
En-NHD-700,8h	300	17.7	-17.7	4.1	-4.2	442	-432
	2	19.4	-19.5	7.5	-7.4	897	-897