

Morphological Studies of Composite Spin Crossover@SiO₂ Nanoparticles

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Table S1. Experimental conditions for the synthesis of samples 1-18.

sample	Temperature (°C)	*V _{H2O} (mL)	ω	*V _{TEOS} (mL)	TEOS inclusion phase	V _{Ammoniac}
1	20	0.5	10	0	---	0
2	20	0.5	10	0.05	organic	0
3	20	0.5	10	0.1	organic	0
4	20	0.5	10	0.2	organic	0
5	0	0.5	10	0.05	organic	0
6	30	0.5	10	0.05	organic	0
7	60	0.5	10	0.05	organic	0
8	20	0.5	10	0.05	aqueous	0
9	20	0.5	10	0.1	aqueous	0
10	20	0.5	10	0.2	aqueous	0
11	20	0.5	10	0.05	starting microemulsions	0
12	20	0.25	5	0.05	starting microemulsions	0
13	20	0.5	10	0.05	final microemulsion	0
14	20	0.5	10	0.05	final microemulsion	0.062
15	20	0.5	10	0.05	final microemulsion	0.124
16	20	0.25	5	0.05	final microemulsion	0
17	20	0.25	5	0.05	final microemulsion	0.062
18	20	0.25	5	0.05	final microemulsion	0.124

* Volume given for each starting microemulsion

Table S2: Elemental analyses and formula for sample 1-18

Compound			C / %	H / %	N / %	B / %	Fe / %	Calculated Formula
1		exptl	19.98	1.62	33.37	2.93	15.29	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{H}_2\text{O})_{0.3}$
			20.02	1.49	33.38	2.81	15.34	
		calcd	20.35	2.43	35.62	3.05	15.69	
2		exptl	19.03	1.53	31.94	2.67	14.16	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.6} \cdot (\text{H}_2\text{O})_{0.5}$
			18.85	1.43	31.77	2.69	14.01	
		calcd	18.30	2.28	32.03	2.74	14.11	
3		exptl	18.03	1.44	30.33	2.42	14.81	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.9} \cdot (\text{H}_2\text{O})_{0.1}$
			18.16	1.60	30.54	2.42	14.78	
		calcd	17.84	2.01	31.23	2.67	13.75	
4		exptl	19.22	1.94	32.08	2.69	14.87	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.5} \cdot (\text{H}_2\text{O})_{0.4}$
			19.24	1.91	32.17	2.59	14.74	
		calcd	18.67	2.28	32.68	2.80	14.39	
5		exptl	19.88	1.71	33.57	2.65	15.72	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.3} \cdot (\text{H}_2\text{O})_{0.4}$
			19.80	1.50	33.26	2.57	15.81	
		calcd	19.38	2.35	33.93	2.90	14.94	
6		exptl	17.21	1.38	28.81	2.44	15.12	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{1.1} \cdot (\text{H}_2\text{O})_{0.4}$
			17.24	1.40	28.88	2.44	14.86	
		calcd	17.03	2.11	29.81	2.55	13.13	
7		exptl	18.90	1.45	31.86	2.72	14.82	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.6} \cdot (\text{H}_2\text{O})_{0.3}$
			18.83	1.48	31.64	2.65	14.70	
		calcd	18.39	2.17	32.19	2.75	14.18	
8		exptl	19.64	1.43	33.05	2.75	15.53	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.3} \cdot (\text{H}_2\text{O})_{0.4}$
			19.70	1.56	33.21	2.65	15.36	
		calcd	19.21	2.34	33.62	2.88	14.81	
9		exptl	19.29	1.61	32.24	2.55	15.71	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.4} \cdot (\text{H}_2\text{O})_{0.5}$
			19.29	1.56	32.23	2.47	15.83	
		calcd	18.82	2.36	32.94	2.82	14.51	
10		exptl	18.27	1.08	29.40	2.74	14.18	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_1 \cdot (\text{H}_2\text{O})_{0.4}$
			18.22	1.14	29.26	2.72	13.83	
		calcd	17.32	2.12	30.31	2.59	13.35	
11		exptl	19.12	1.50	32.57	3.12	14.94	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.5} \cdot (\text{H}_2\text{O})_{0.4}$
			19.01	1.44	32.39	3.20	15.23	
		calcd	18.79	2.29	32.88	2.81	14.48	
12		exptl	18.32	1.43	31.12	3.00	15.59	$[\text{Fe}(\text{Htrz})_2(\text{Trz})](\text{BF}_4) \cdot (\text{SiO}_2)_{0.6} \cdot (\text{H}_2\text{O})_{0.6}$
			18.28	1.46	31.11	2.93	15.34	
		calcd	18.16	2.32	31.79	2.72	14.00	
13		exptl	18.28	1.48	28.74	2.82	14.23	

			18.40	1.75	28.97	2.83	14.12	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{1.1} ·(H ₂ O) _{0.4}
		calcd	17.08	2.08	29.89	2.56	13.16	
14		exptl	16.37	1.38	27.12	2.30	12.22	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{1.6} ·(H ₂ O) _{0.5}
			17.01	1.33	28.24	2.30	12.20	
		calcd	15.98	1.99	27.98	2.39	12.32	
15		exptl	17.35	2.41	19.52	1.97	12.50	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{2.7} ·(H ₂ O) _{1.2}
			17.52	2.57	19.81	2.01	12.89	
		calcd	13.24	1.91	23.16	1.98	10.20	
16		exptl	17.06	1.58	26.08	2.62	13.40	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{1.7} ·(H ₂ O) _{0.5}
			17.08	1.39	26.33	2.53	13.06	
		calcd	15.68	1.95	27.45	2.35	12.09	
17		exptl	15.11	1.38	22.46	2.23	12.67	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{2.7} ·(H ₂ O) _{0.7}
			14.92	1.37	22.23	2.25	12.70	
		calcd	13.76	1.79	24.09	2.06	10.61	
18		exptl	13.34	2.01	19.14	1.62	13.45	[Fe(Htrz) ₂ (Trz)](BF ₄)·(SiO ₂) _{3.6} ·(H ₂ O) _{1.2}
			13.31	2.08	19.12	1.61	13.60	
		calcd	12.28	1.77	21.50	1.84	9.47	

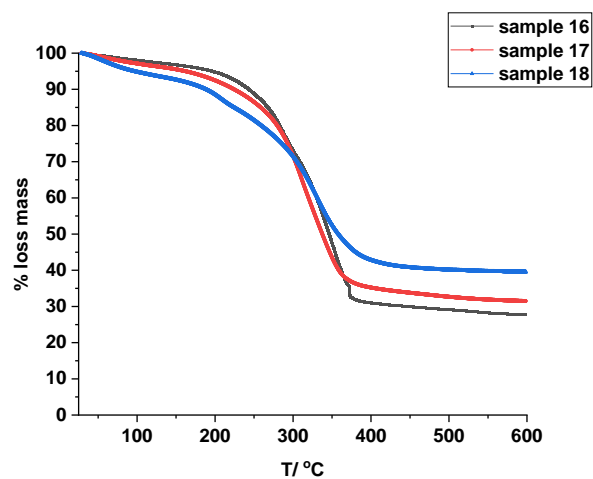
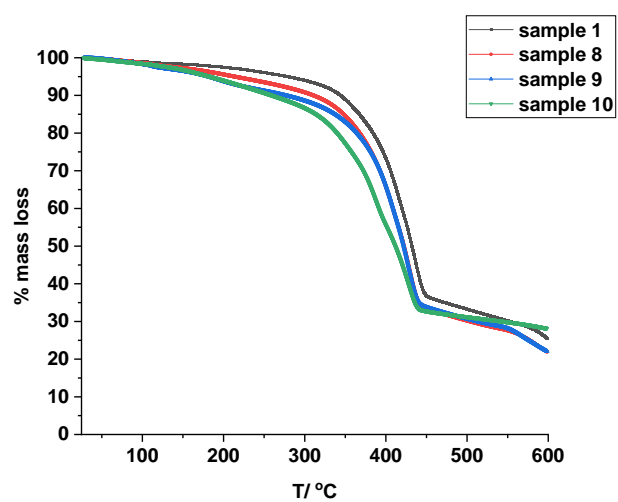
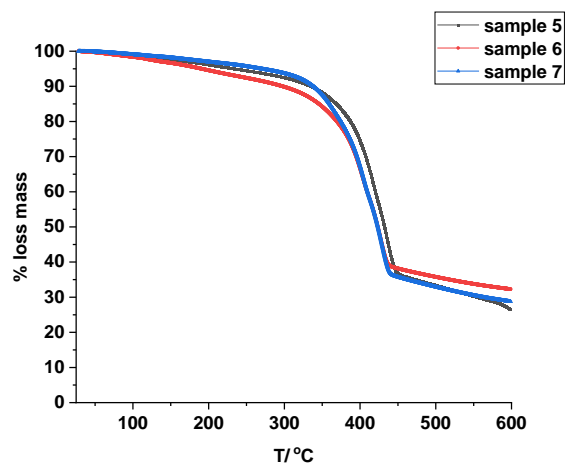
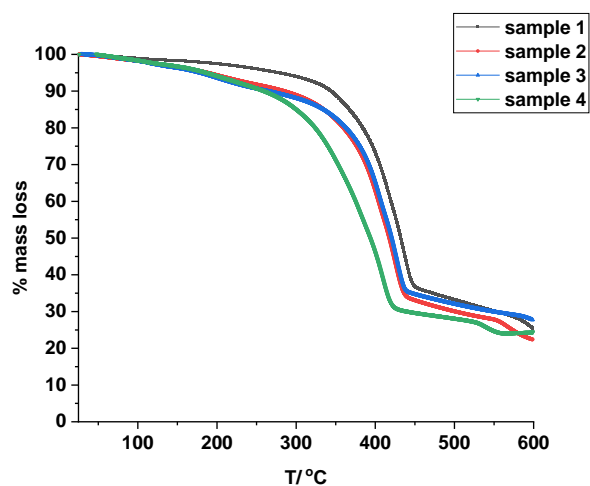
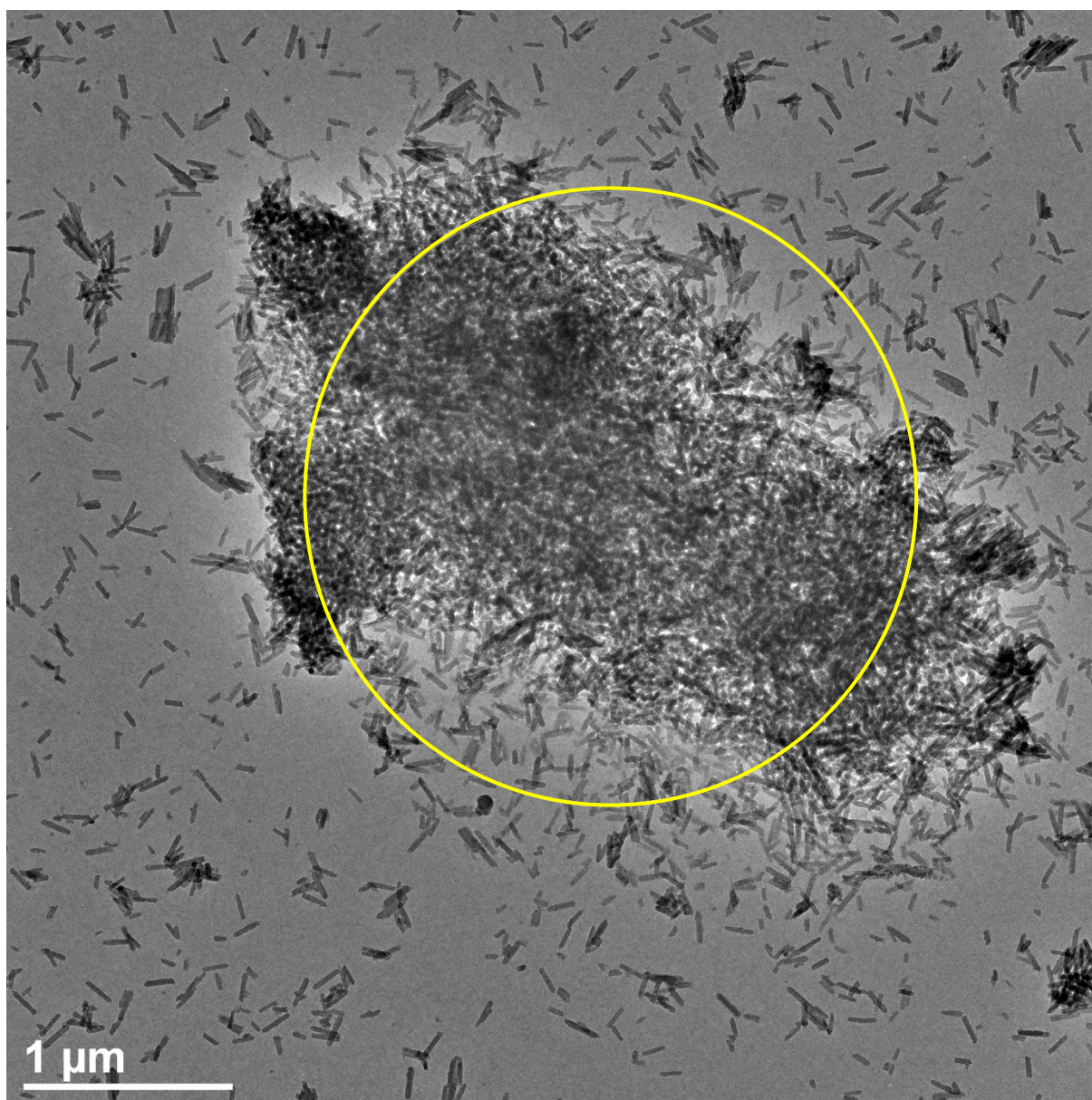


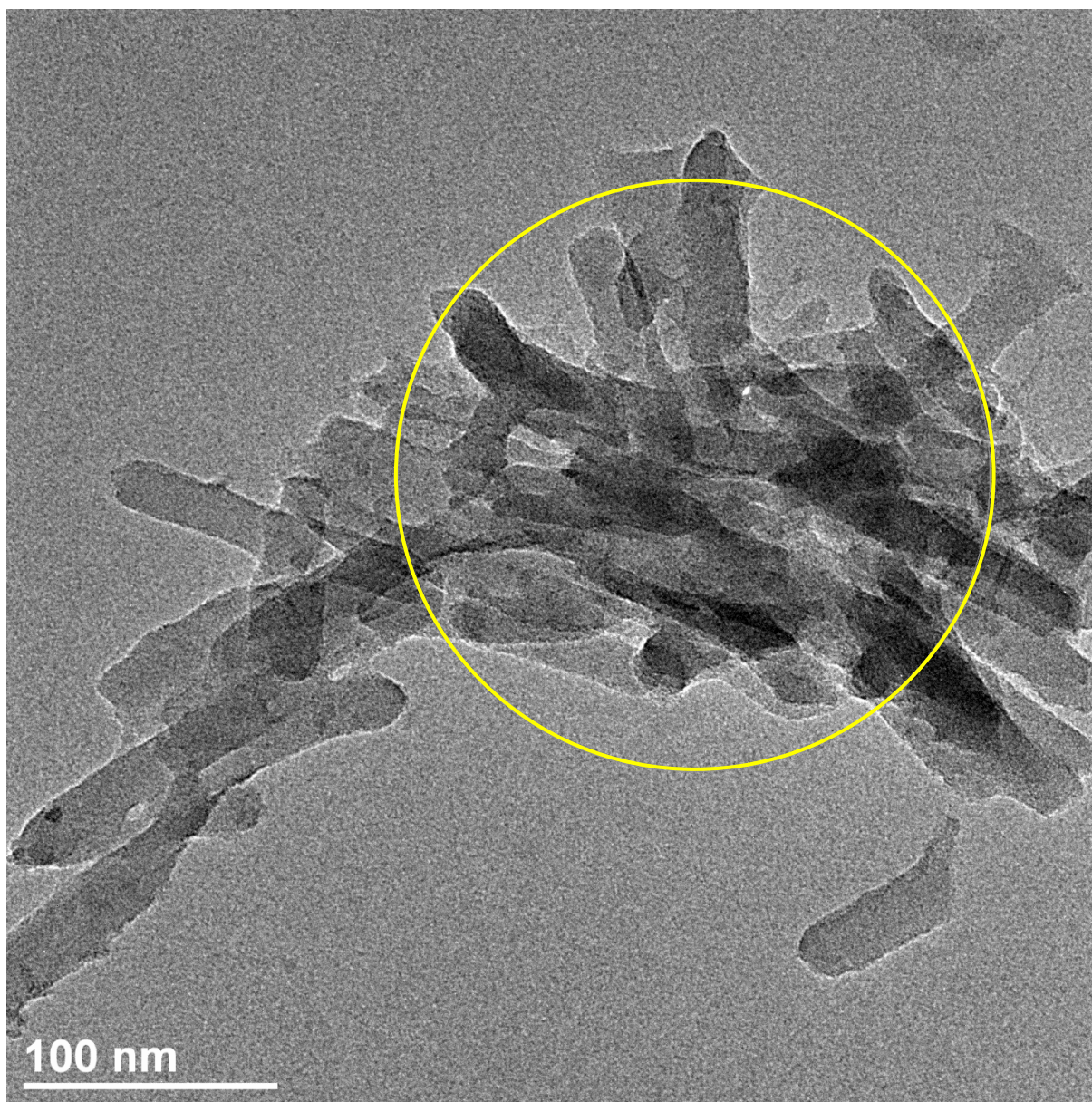
Figure S1: Thermogravimetric analyses

TEM images of sample 2



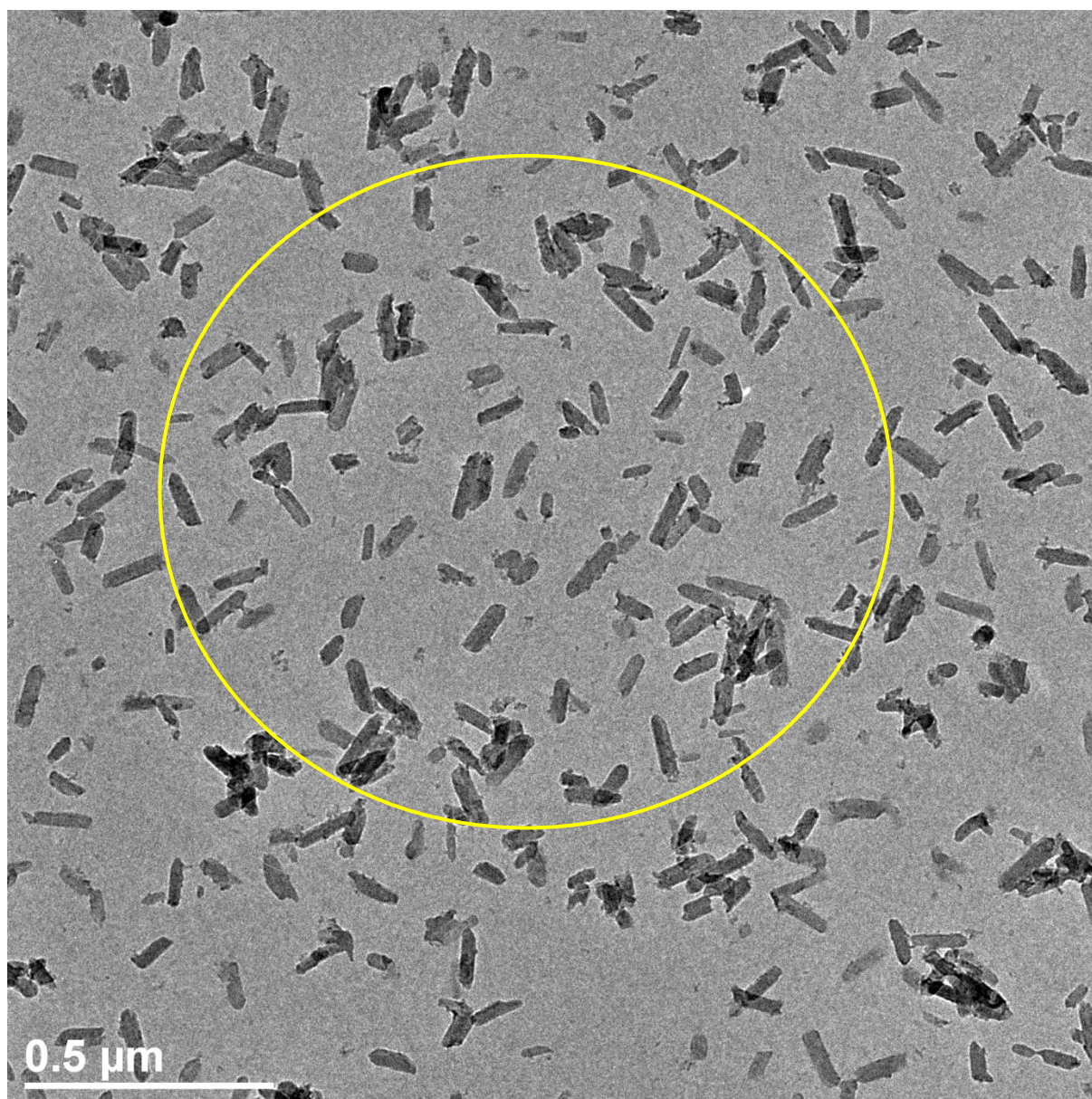
Yellow circle shows the measured area

Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Silicon	K-series	14.91	25.84	0.21
Iron	K-series	85.09	74.16	2.63
Total:		100.00	100.00	

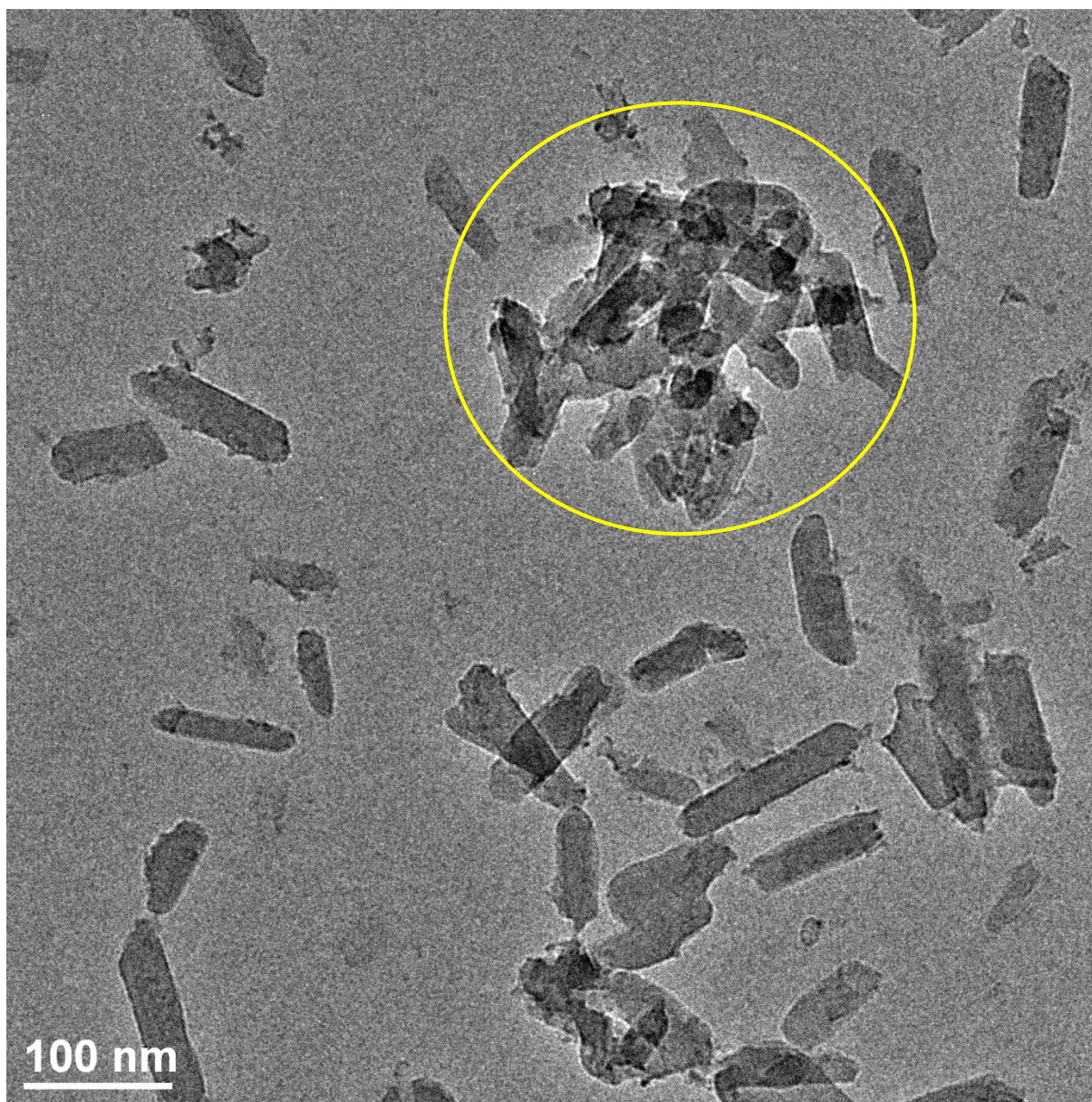


Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Silicon	K-series	16.19	27.76	0.22
Iron	K-series	83.81	72.24	2.60
Total:		100.00	100.00	

TEM images of sample 3

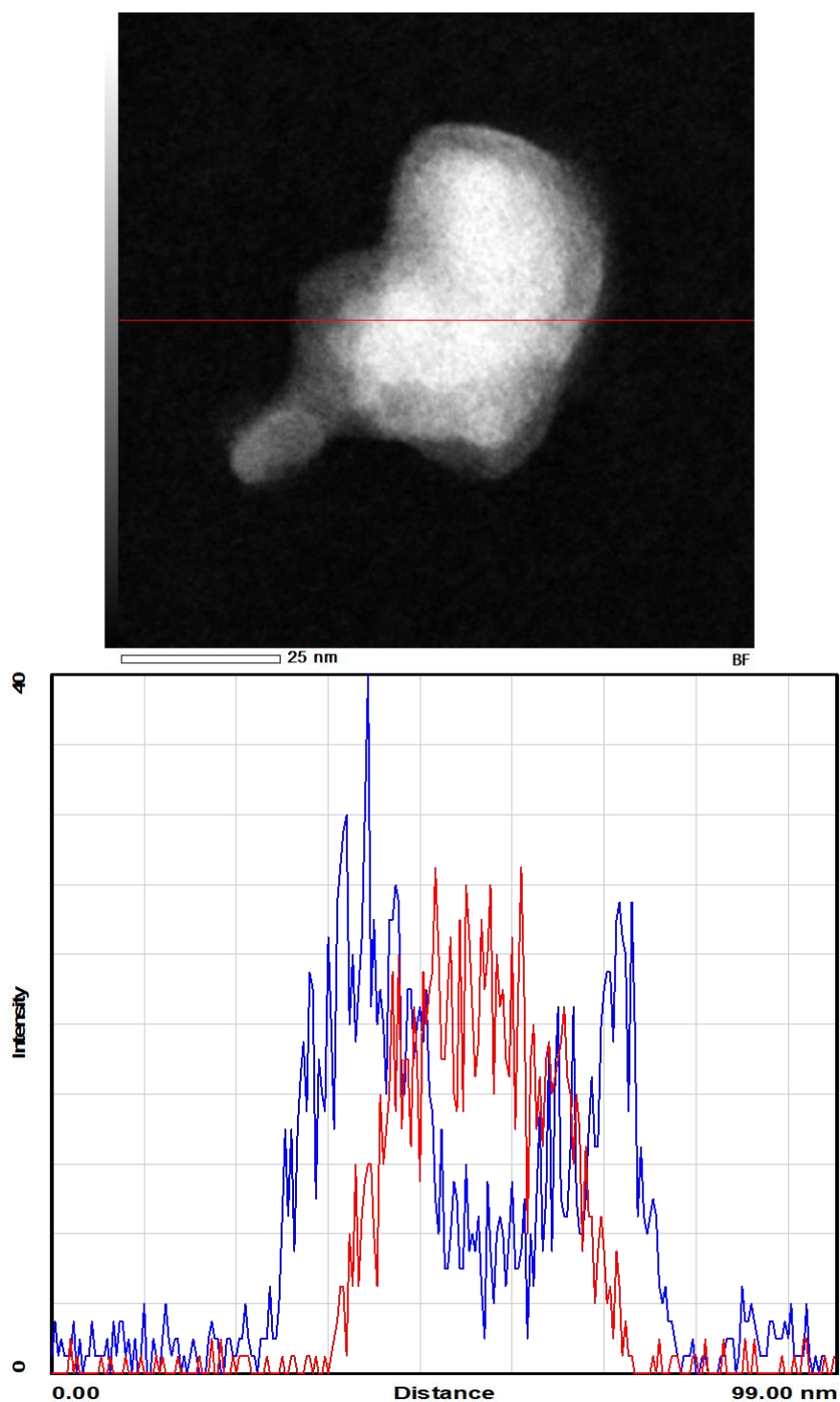


Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Iron	K-series	76.25	61.76	3.37
Silicon	K-series	23.75	38.24	1.17
Total:		100.00	100.00	

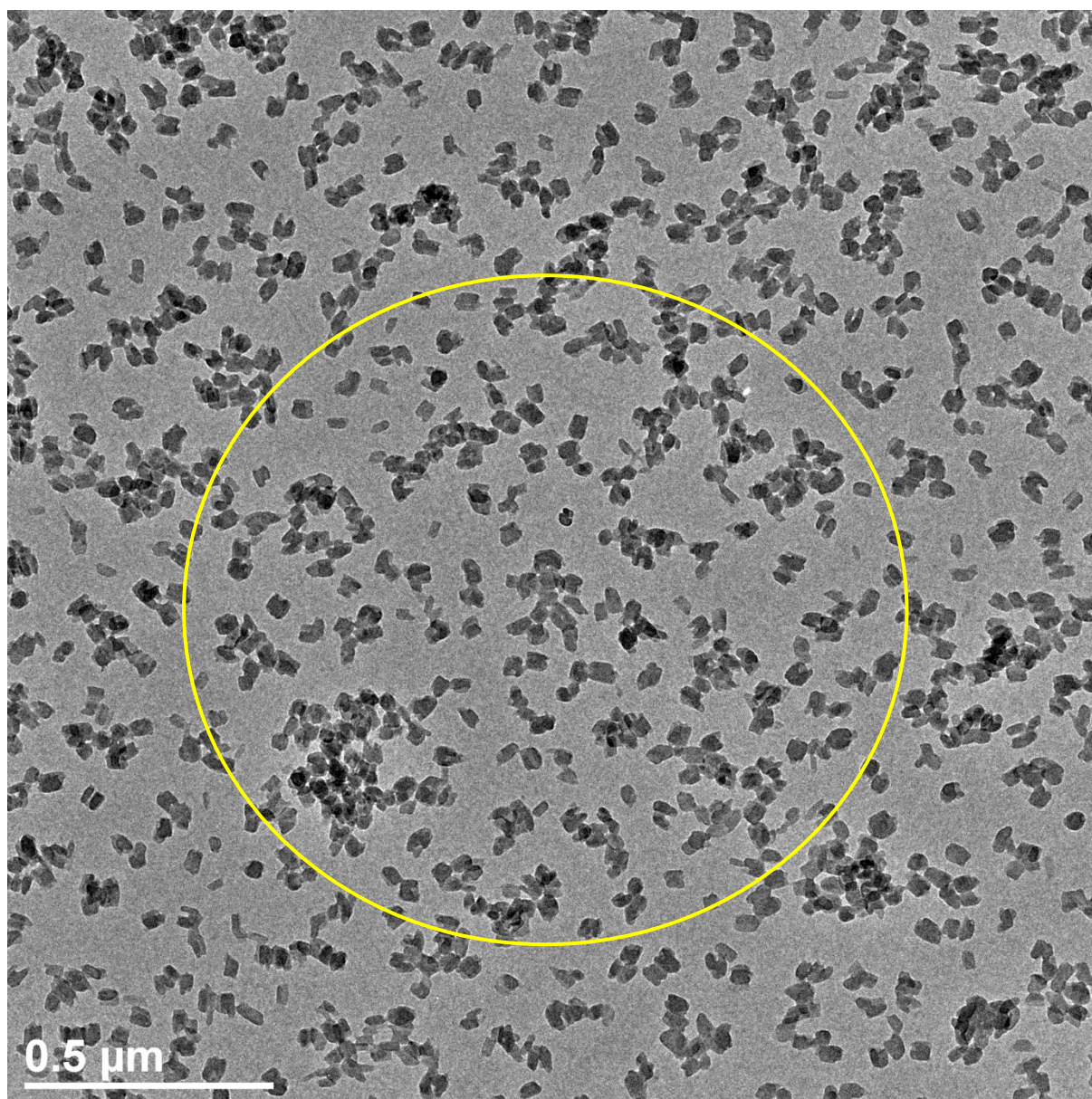


Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Iron	K-series	75.42	60.67	2.68
Silicon	K-series	24.58	39.33	0.69
Total:		100.00	100.00	

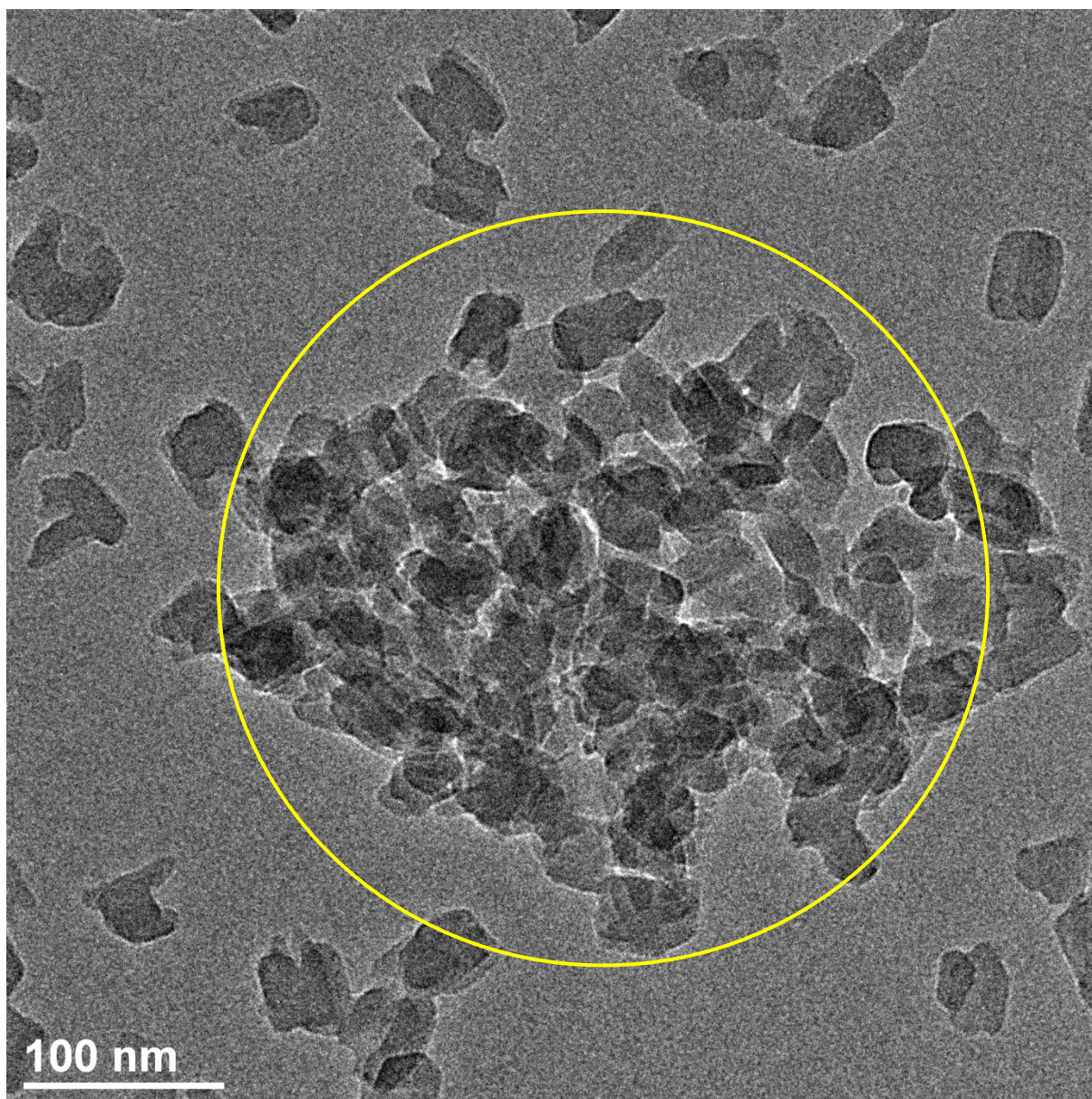
Bright field HRSTEM images and the corresponding EDX distribution of Fe (in red) and Si (in blue) along cross-section line for core@shell nanoparticle sample 3.



TEM images of sample 4

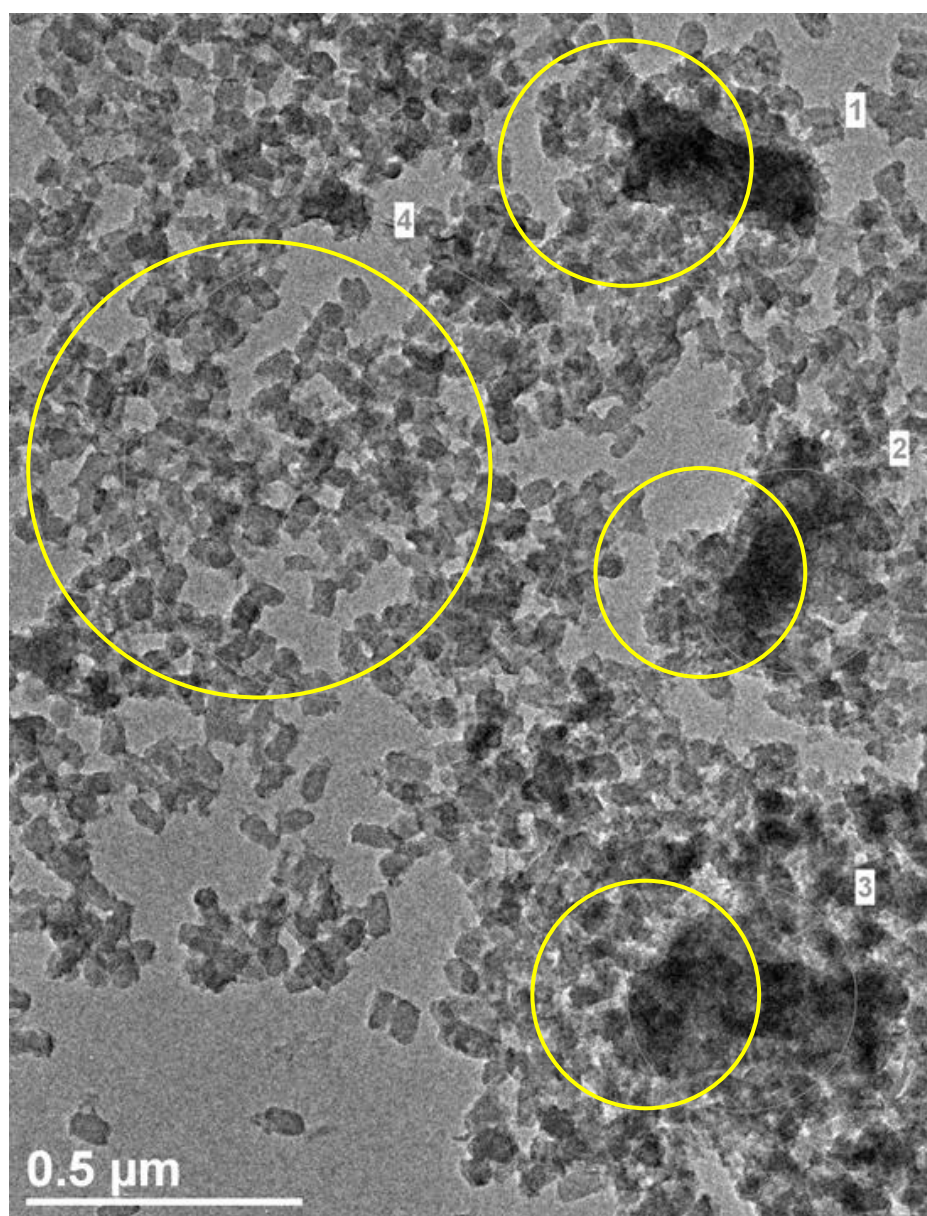


Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Iron	K-series	77.75	63.73	2.60
Silicon	K-series	22.25	36.27	0.52
Total:		100.00	100.00	



Element	Series	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
Iron	K-series	86.60	76.47	2.65
Silicon	K-series	13.40	23.53	0.16
Total:		100.00	100.00	

TEM images of sample 15



spot 3

Element	Series	norm. C [wt.%]	Atom. C [at.%]
Silicon	K-series	29.63	45.57
Iron	K-series	70.37	54.43
Total:		100.00	100.00

spot 4

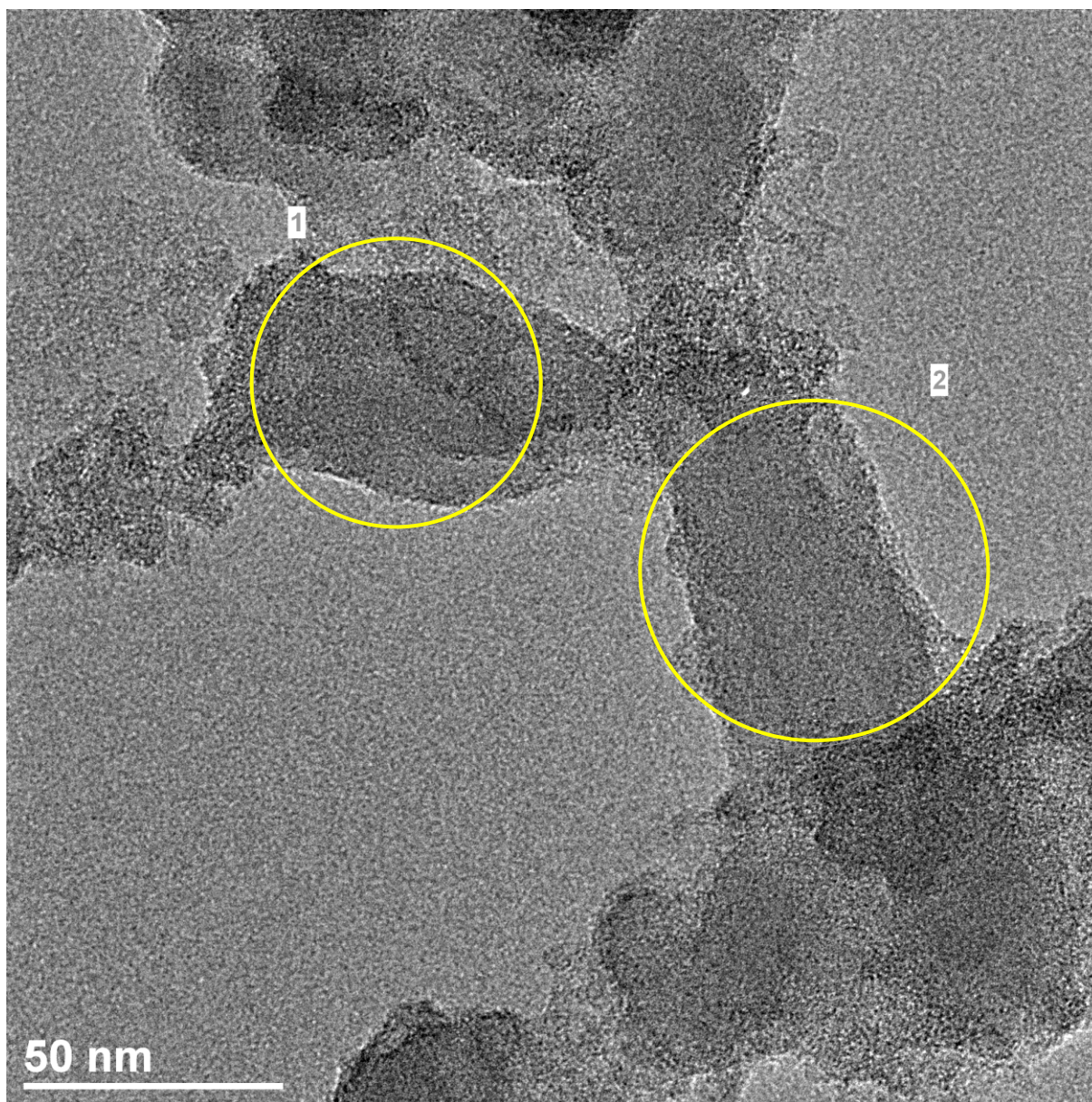
Element	Series	norm. C [wt.%]	Atom. C [at.%]
Silicon	K-series	35.05	51.76
Iron	K-series	64.95	48.24
Total:		100.00	100.00

spot 1

Element	Series	norm. C [wt.%]	Atom. C [at.%]
Silicon	K-series	30.53	46.63
Iron	K-series	69.47	53.37
Total:		100.00	100.00

spot 2

Element	Series	norm. C [wt.%]	Atom. C [at.%]
Silicon	K-series	31.56	47.83
Iron	K-series	68.44	52.17
Total:		100.00	100.00



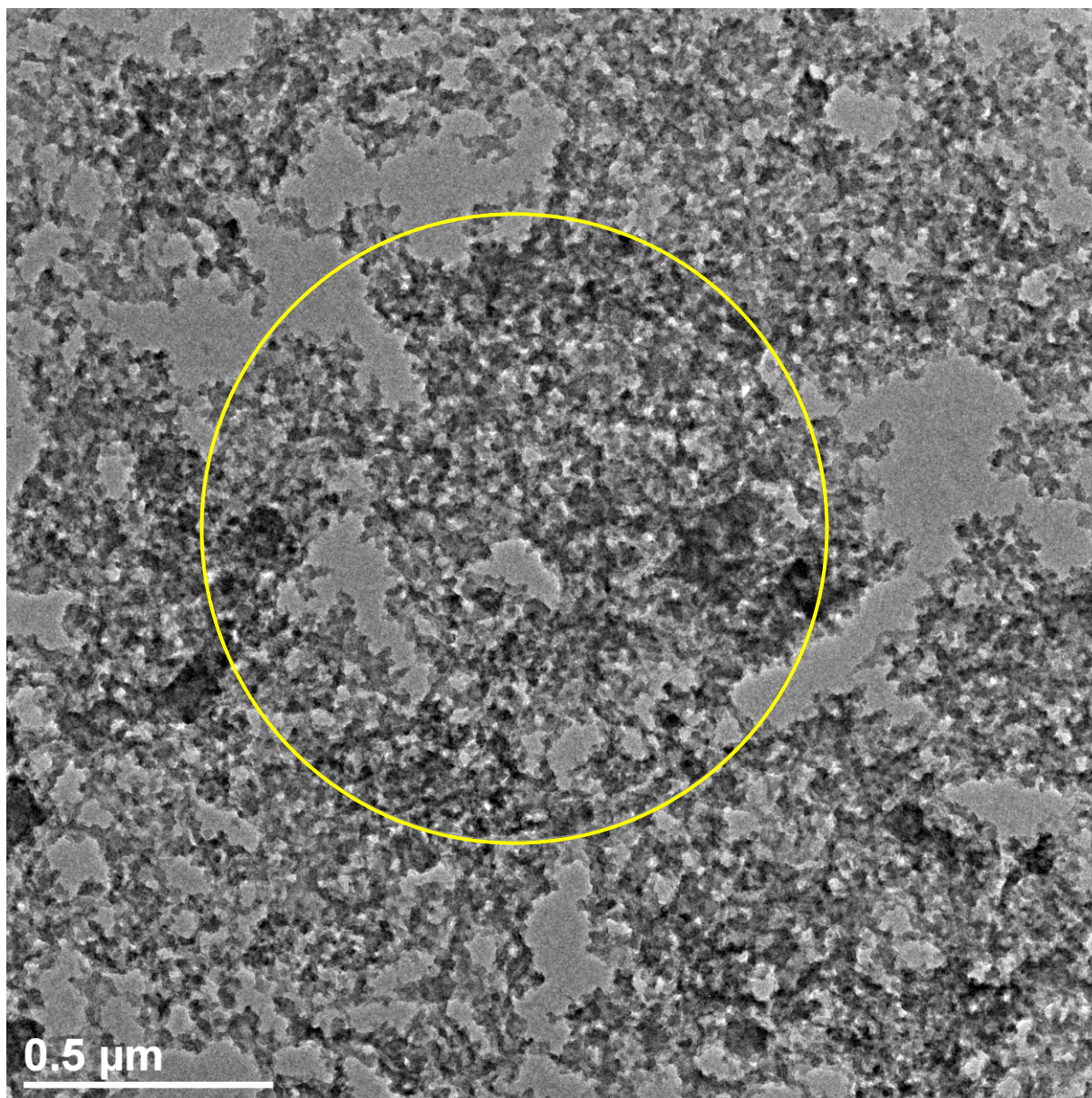
spot 1

Element	Series	norm. C [wt.%]	Atom. C [at.%]
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Silicon	K-series	28.54	44.27
Iron	K-series	71.46	55.73
<hr/>			
Total:		100.00	100.00

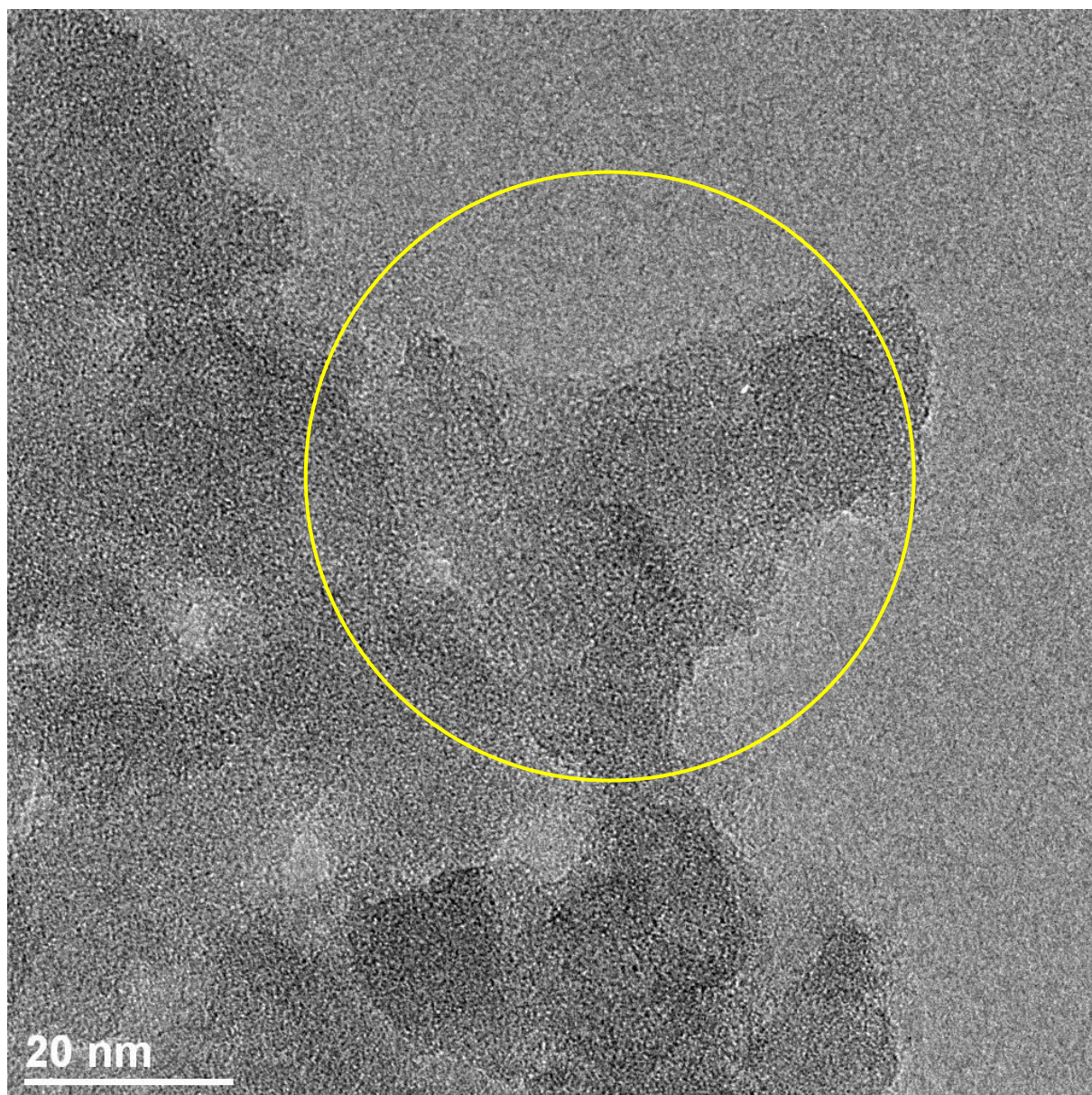
spot 2

Element	Series	norm. C [wt.%]	Atom. C [at.%]
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Silicon	K-series	30.58	46.69
Iron	K-series	69.42	53.31
<hr/>			
Total:		100.00	100.00

TEM images of sample 18



Element	Series	norm. C [wt.%]	Atom. C [at.%]
Iron	K-series	60.58	43.59
Silicon	K-series	39.42	56.41
Total:		100.00	100.00



Element	Series	norm. C [wt.%]	Atom. C [at.%]
<hr/>			
Silicon	K-series	48.29	65.00
Iron	K-series	51.71	35.00
<hr/>			
Total:		100.00	100.00

Figure S2: Transmission Electronic Microscopy (TEM) and EDX analyses

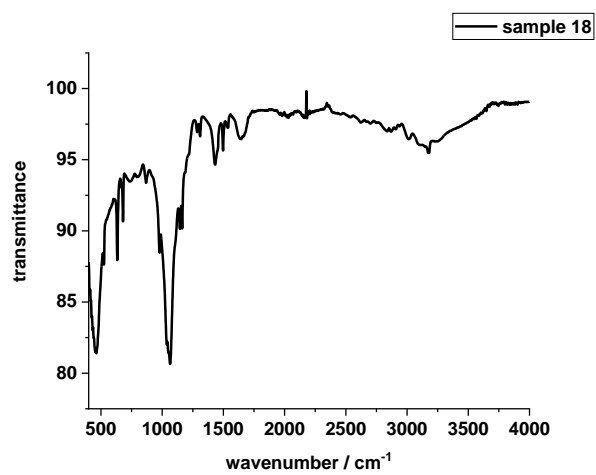
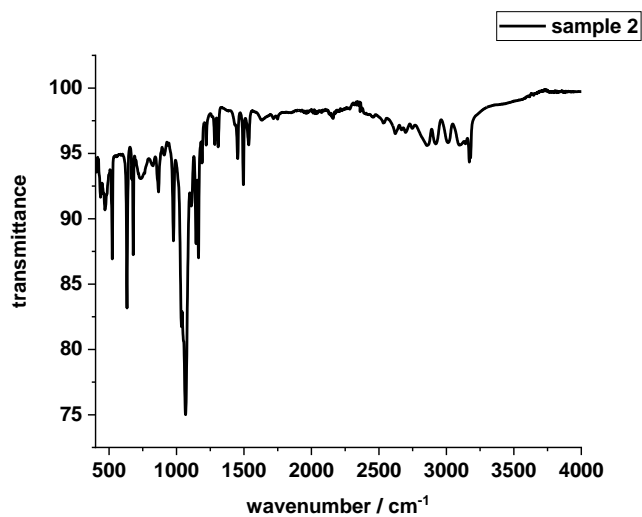
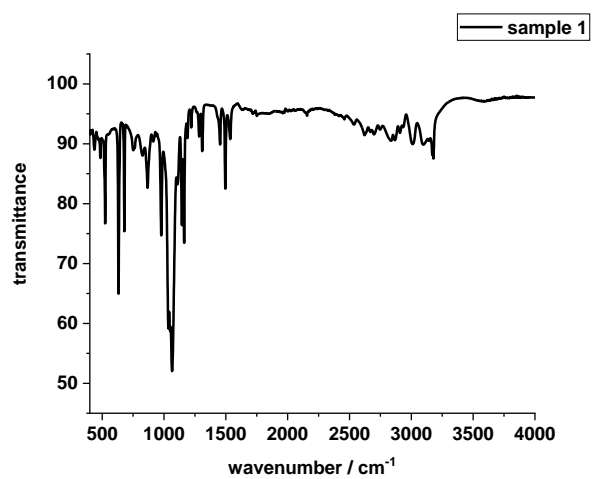
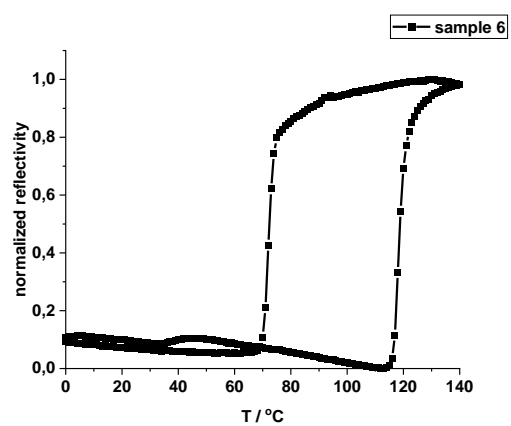
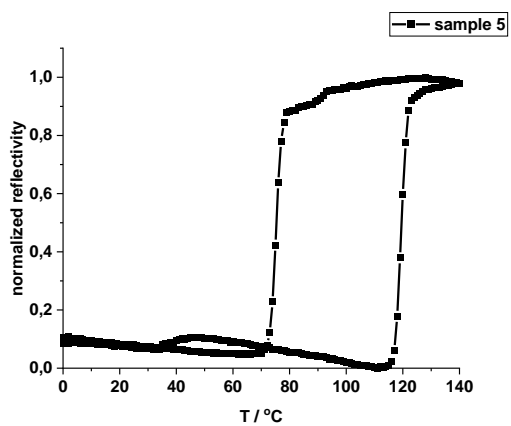
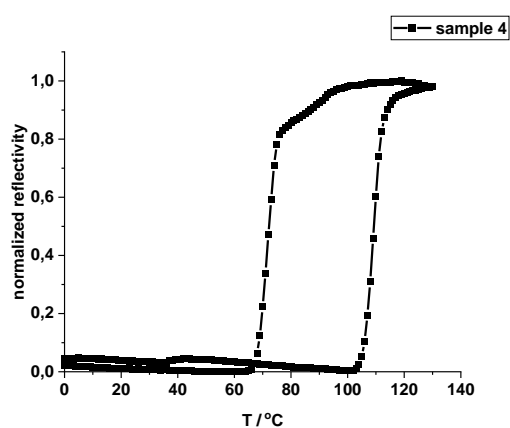
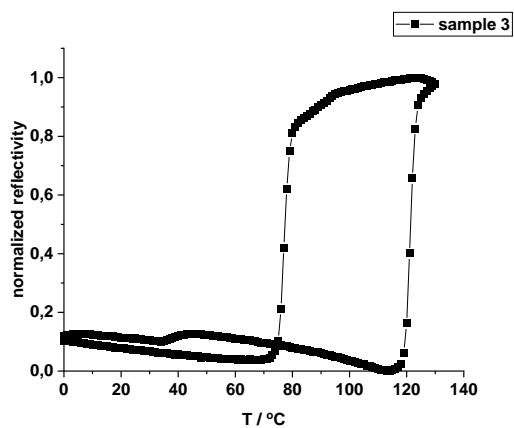
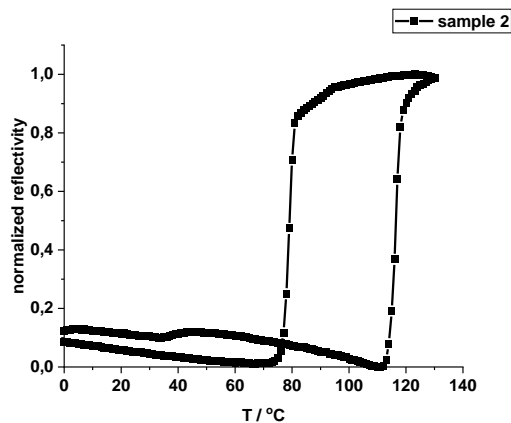
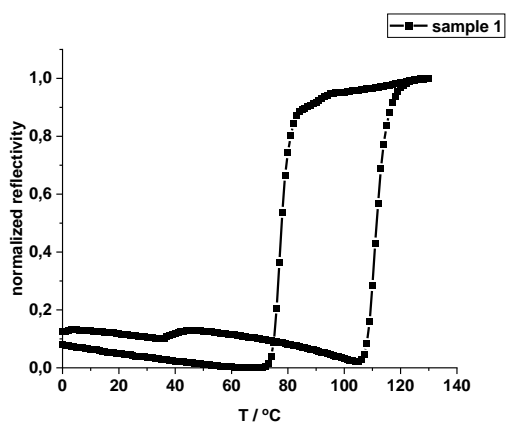
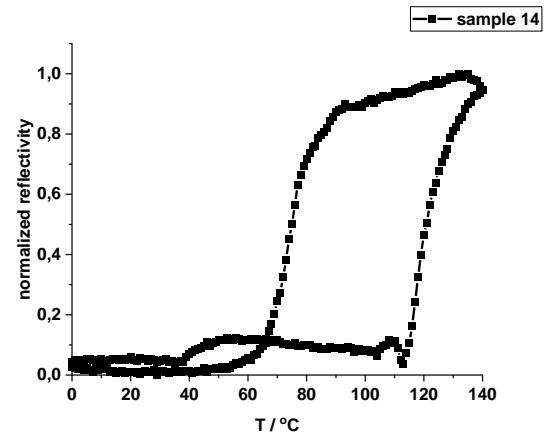
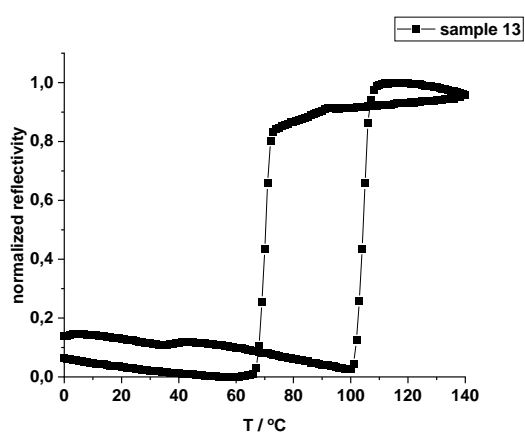
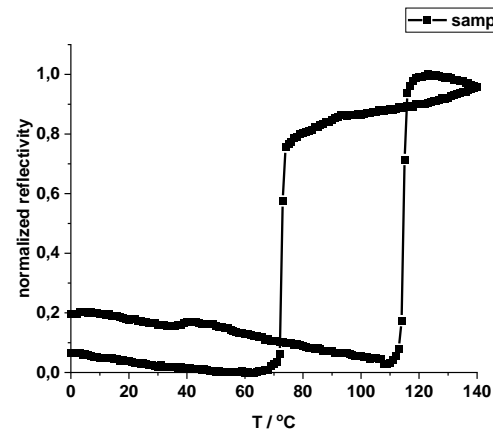
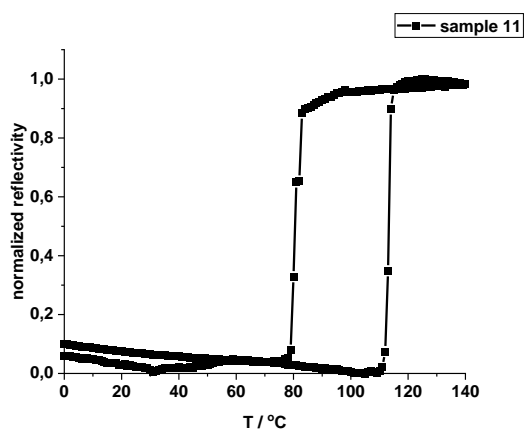
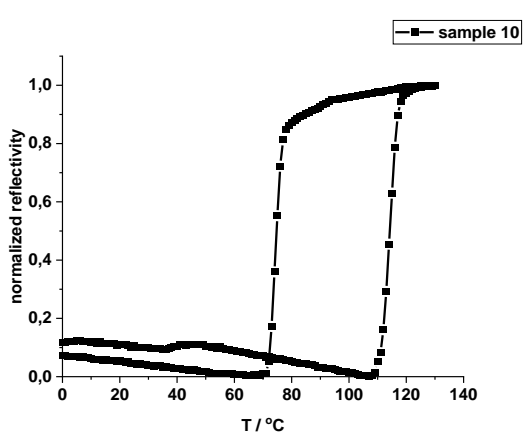
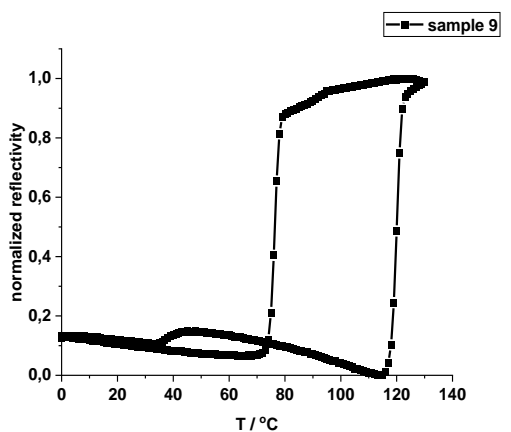
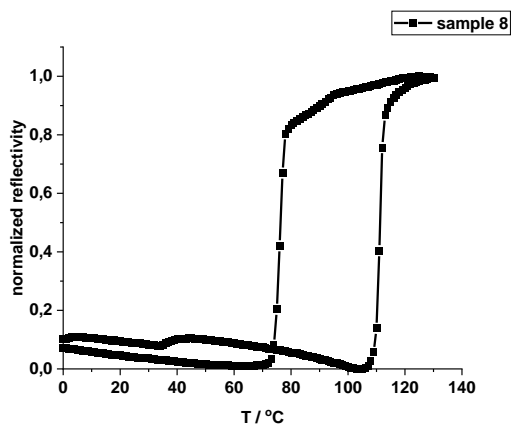
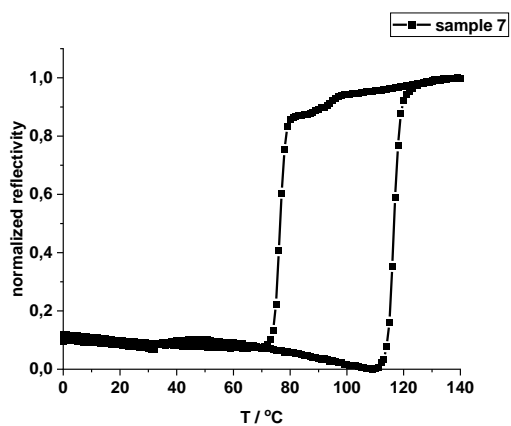


Figure S3: Infrared spectroscopy





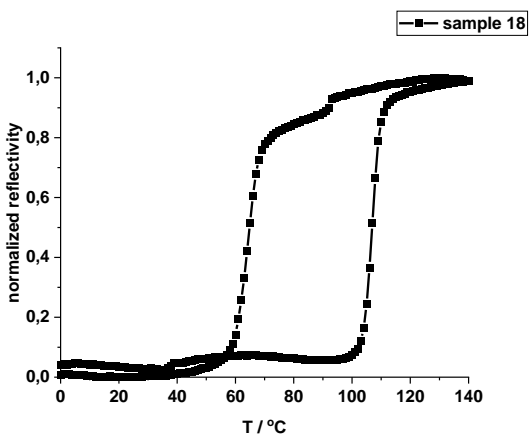
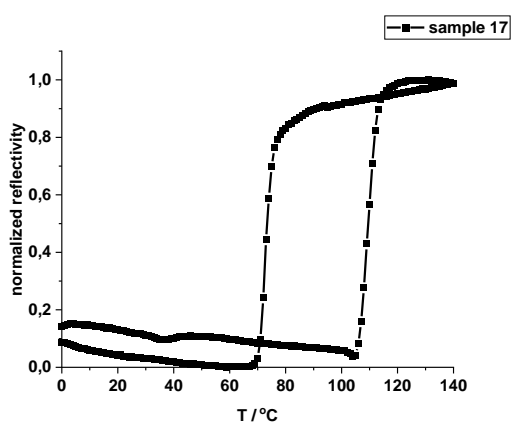
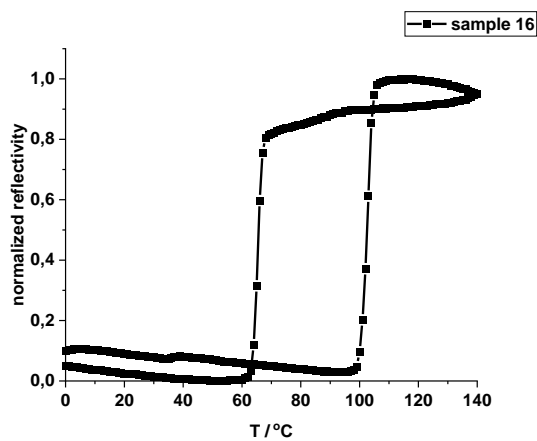
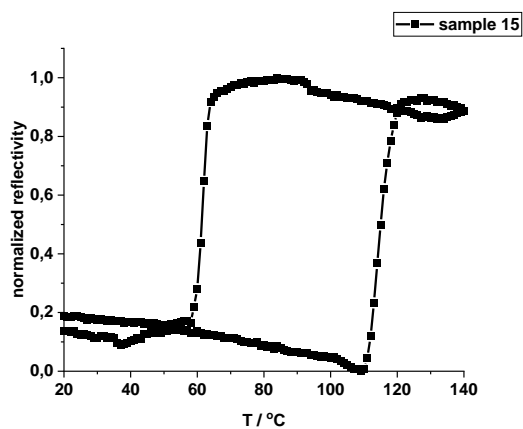


Figure S4: Optical reflectivity measurements for samples 1-18