

# Reproducibility and Scalability of Magnetic Nanoheater Synthesis

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## Experimental procedures for the characterization of the MNP's.

TEM observations were made using a JEOL JEM 1011 transmission electron microscope operated at 100 kV with Gatan ES1000Ww camera. Samples were prepared by placing one drop of a dilute particle suspension in toluene on an amorphous carbon-coated copper grid (Carbon coating system Leica EM MED020) and evaporation the toluene at room temperature. In average approximately a thousand of particles were manually counted on 3-5 microphotographs, but in the more polydisperse samples in those more microphotographs were studied given the tendency of smaller and bigger nanoparticles to get together between them during drying.

To obtain dry samples, the dispersion of MNP's in Oleic acid: Toluene 1:7 v/v (3 mL) was precipitated using methanol (10 mL). After the elimination of the supernatant, the MNP's were washed one time with methanol (10 mL) and dried at 50°C. These samples were used for XRD, TG-ATD and magnetometry.

XRD was performed in a Bruker D8 Advance DAVINCI diffractometer using a zero background holder for the sample (PANalytical 32mm diameter silicon insert adapted for its use in our diffractometer). The analysis was performed in the range of 20-49° in 2  $\theta$  with a scan step size of 0.01° and a scan step time of 1s. The Scherrer size was determined for the three more intense magnetite peaks (220), (311) and (400) using the EVA software.

Thermogravimetric analysis was performed simultaneously in a TG/DTA Q600 TA Instruments. In this work the sample (approximately 10 mg) was heated in Air (100 ml/min) until 800 °C at a scanning rate 10 °C/min. As usual the weight loss between 200 and 800 °C was considered as the organic content of the sample.

Magnetic characterization of the samples was carried out in a vibrating sample magnetometer MagLabVSM (Oxford Instruments) with a maximum field of 5 T. Hysteresis loops of the powder samples were measured at room temperature at a rate of 0.3 T/min. The saturation magnetization was determined by extrapolating to infinite field the magnetization data above 2.5 T using the linear  $M(H)$  vs  $1/H$  plot. The magnetic susceptibility was determined as the slope of the  $M$  vs  $H$  below 0.02T where the dependence of the magnetization and the magnetic field is linear. The mass magnetization was presented in emu/g and the magnetic susceptibility as emu/g T.

The dispersion samples (1 mL) for the measurements of the solvodynamic size  $D_{SDY}$  by PCS and SAR by AMF heating measurements were ultrasonically dispersed using an Elmasonic S30 in a glass test tube for 5x2 min and measured immediately.

## Comparison among the Scaled and the standard experiment

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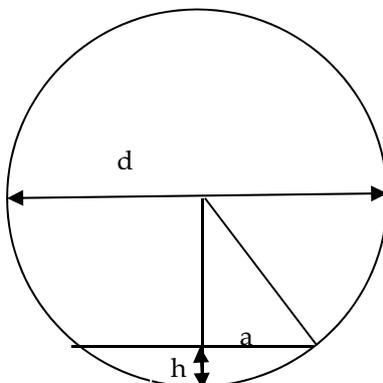
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### 1-. Energy Flux

We have made measurements of the height (h) that the reactant mixture reaches inside the two spherical reactors of the synthesis and of the internal diameter (d) to calculate the surface A (area of the spherical cap) in contact to the reactant through which the energy is transported.



$$a = \sqrt{r^2 - (r - h)^2}$$

$$A = \pi \cdot (a^2 + h^2)$$

The power that is delivered through the spherical cap (w) is approximated by the total power supplied (300 W for the 1 L reactor, and 1300 W for the 10 L reactor) multiplied by the ratio between its surface A and that of the hemisphere S. The energy flow ( $\phi$ ) will finally be obtained by dividing w by A.

reactor	d [cm]	h [cm]	r [cm]	a [cm]	A [cm <sup>2</sup> ]	S [cm <sup>2</sup> ]	A/S	w [W]	$\Phi$ [W/cm <sup>2</sup> ]
1 L	13	1.8	6.5	4.49	73.5	265	0.277	83.1	<b>1.13</b>
10 L	27.5	4.6	13.75	10.26	397	1187	0.334	434.2	<b>1.09</b>

There are no major differences in terms of energy flow, if anything it is less in the scaled experiment.

### 2-. Energy delivered per gram of reactant.

The energy received per gram of reagent considering 61.31 g of reagent from the 1 L experiment and 1186 g from the 10 L experiment would be  $83.1 / 61.31 = 1.36 \text{ W / g}$  for the 1 L experiment and  $434.2 / 1186 = 0.366 \text{ W / g}$  for the one with 10 L. Therefore, the increase in size of the particles when scaled is not due to having more energy available.

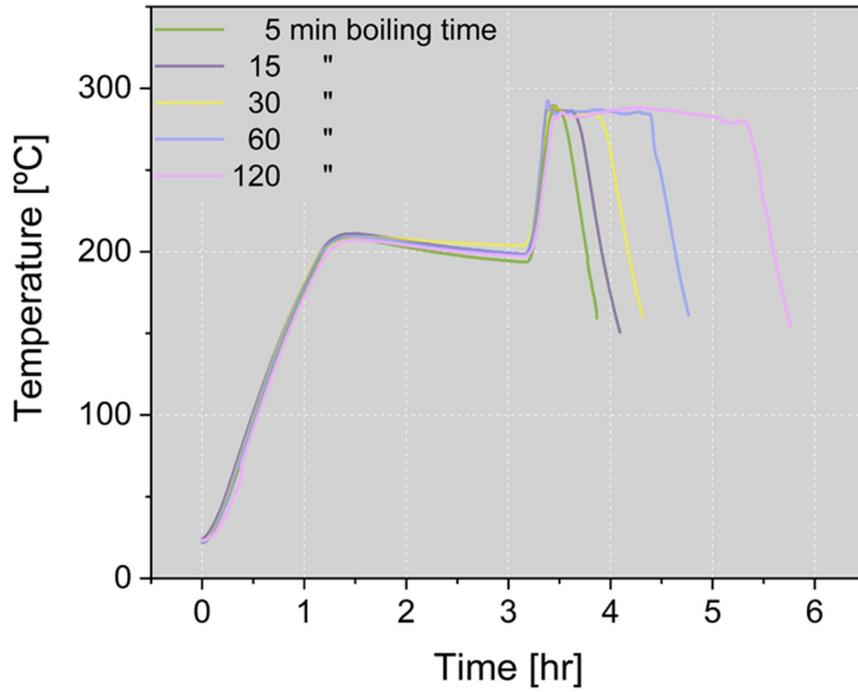


Figure S1 Temperature profiles for the scaled-up experiments

Table S1 Average parameters of the temperature profiles for each boiling time, averages of all the experiments and their repeatability

Boiling time [min]	Ramp 1 25-200 °C [°C/min]	Plateau 200°C		Ramp 2 200-288 °C [°C/min]	Boiling temperature	
		$\bar{T}$ [°C]	$\sigma_T$ [°C]		$\bar{T}$ [°C]	$\sigma_T$ [°C]
5	2.76±0.04	201.1±1.4	4.7±0.5	9.04±0.23	285.8±3.8	2.1±0.6
15	2.75±0.03	204.7±3.1	4.3±1.4	9.22±0.12	286.9±2.1	1.9±0.5
30	2.83±0.06	206.1±4.8	2.3±0.8	8.52±0.26	284.8±1.3	2.5±0.4
60	2.80±0.01	203.9±4.7	3.7±1.0	9.06±0.62	285.5±0.6	2.0±1.3
120	2.71±0.07	202.1±1.1	3.5±0.4	9.29±0.39	284.4±1.3	3.5±0.7
<b>Global Averages</b>	<b>2.77±0.04</b>	<b>203.6±1.6</b>	<b>3.7±0.7</b>	<b>9.03±0.24</b>	<b>285.5±0.8</b>	<b>2.4±0.5</b>
$\sigma$	<b>0.041</b>	<b>1.79</b>	<b>0.82</b>	<b>0.27</b>	<b>0.87</b>	<b>0.59</b>
<b>Reproducibility (1-<math>\sigma/\bar{x}</math>) · 100</b>	<b>98.5%</b>	<b>99%</b>	<b>77.8%</b>	<b>97%</b>	<b>99%</b>	<b>75.4%</b>

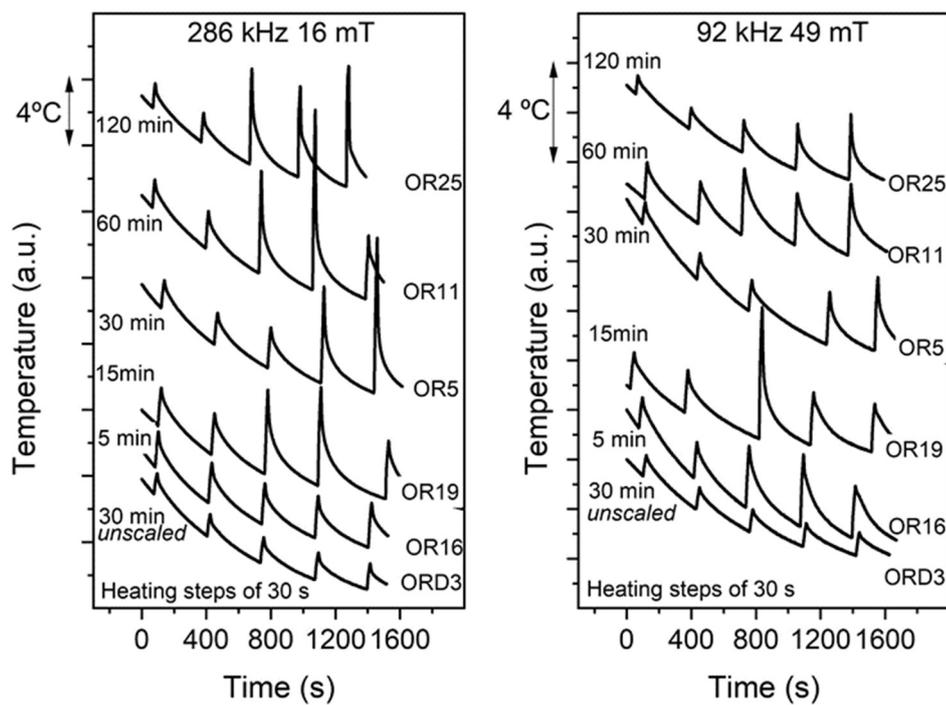
**TABLE S2** Experimental data of scaled-up samples. Reproducibility  $r = (1 - \sigma/\bar{x}) \cdot 100$ . Error of the average:  $1.96 \cdot \sigma/\sqrt{N}$  (95% confidence)  $N=6$

	Exp.	Yield [%]	TEM			DRX Scherrer's size	PCS $D_{SDV}$ 0.1 mgFe/mL		TG-ATD	Magnetic properties at room temperature		MAGNETIC HIPERTHERMIA 1mgFe/mL	
			$D_{TEM}$ [nm]	$\sigma_{TEM}$ [nm]	$\sigma_{TEM}/D_{TEM}$	$D_{DRX}$ [nm]	$Z_{ave}$ [nm]	PDI	Organic wt%	Ms emu/g	$\gamma$ emu/gT	286 kHz $18 \text{ kAm}^{-1}$	92 kHz $50 \text{ kAm}^{-1}$
													SAR [W/gFe]
5 min	OR1 2	36	9.0	2.5	0.28	13.2±0.6	170	0.8	8.35	66.43±0.19	847±51	100	58
	OR1 3	35	12.8	3.5	0.27	12.6±1.8	43	0.3	9.40	65.39±0.51	794±46	109	85
	OR1 4	13	15.2	6.0	0.39	13.1±0.8	56	0.4	11.28	64.44±0.45	1004±62	159	101
	OR1 5	61	11.5	4.7	0.41	13.0±1.2	58	0.4	8.87	73.15±1.1	1188±69	164	57
	OR1 6	38	13.6	4.5	0.33	13.2±1.8	51	0.3	8.85	67.74±0.43	923±42	155	80
	OR3 0	40	15.3	4.1	0.27	12.7±0.5	38	0.2	10.94	67.76±0.27	873±47	163	62
$\bar{x}$		<b>37±11</b>	<b>13±2</b>	<b>4.2±0.9</b>	<b>0.33±0.05</b>	<b>13.0±0.2</b>	<b>69±36</b>	<b>0.4±0.2</b>	<b>9.6±0.9</b>	<b>67.5±0.2</b>	<b>938±103</b>	<b>142±22</b>	<b>74±13</b>
$\sigma$		<b>14</b>	<b>2.2</b>	<b>1.1</b>	<b>0.057</b>	<b>0.23</b>	<b>45</b>	<b>0.2</b>	<b>1.10</b>	<b>2.8</b>	<b>129</b>	<b>27</b>	<b>16.2</b>
r [%]		<b>62</b>	<b>83</b>	<b>74</b>	<b>83</b>	<b>98</b>	<b>35</b>	<b>50</b>	<b>88</b>	<b>96</b>	<b>86</b>	<b>81</b>	<b>78</b>
15 min	OR1 7	71	15.5	4.2	0.27	13.8±1.1	62	0.5	9.15	65.52±0.16	1105±60	229	100
	OR1 8	82	12.9	4.0	0.31	13.1±1.3	63	0.6	8.82	65.91±0.25	923±46	177	86
	OR1 9	40	15.7	5.6	0.36	14.0±1.1	77	0.8	9.90	64.54±0.22	709±30	155	81
	OR2 0	30	13.1	4.7	0.36	12.9±1.2	52	0.4	8.34	64.79±0.11	813±49	128	91
	OR2 1	21	13.6	4.0	0.29	12.7±0.7	66	0.5	8.82	62.37±0.13	998±57	113	70
	OR3 1	44	13.4	3.8	0.29	13.2±1.9	144	0.5	9.42	68.49±0.06	885±41	109	54
$\bar{x}$		<b>48±18</b>	<b>14±0.8</b>	<b>4.4±0.5</b>	<b>0.31±0.03</b>	<b>13.3±0.4</b>	<b>77±24</b>	<b>0.6±0.1</b>	<b>9.1±0.4</b>	<b>65.27±1.5</b>	<b>906±101</b>	<b>152±34</b>	<b>80±12</b>
$\sigma$		<b>22</b>	<b>1.1</b>	<b>0.6</b>	<b>0.035</b>	<b>0.47</b>	<b>30</b>	<b>0.12</b>	<b>0.50</b>	<b>1.82</b>	<b>127</b>	<b>42</b>	<b>15</b>
r [%]		<b>54</b>	<b>92</b>	<b>86</b>	<b>89</b>	<b>96</b>	<b>61</b>	<b>80</b>	<b>95</b>	<b>97</b>	<b>86</b>	<b>72</b>	<b>81</b>
30 min	OR1	57	11.5	3.9	0.34	13.1±1.7	220	0.8	10.91	59.59±0.18	665±24	105	70
	OR2	51	16.7	6.0	0.36	13.6±1.4	108	0.8	11.35	62.78±0.24	817±31	150	77
	OR3	81	10.4	4.1	0.39	11.5±2.5	70	0.8	15.01	60.01±0.09	738±39	98.2	61
	OR5	55	10.0	3.4	0.34	14±0.57	123	0.6	7.33	63.69±0.13	760±26	111	67
	OR2 2	69	15.9	5.8	0.36	13.5±1.4	40	0.3	8.98	65.49±0.23	824±45	100	69
	OR2 3	21	14	5	0.36	14.3±1.4	61	0.6	10.55	66.67±0.34	1063±74	175	81
$\bar{x}$		<b>56±16</b>	<b>13±2</b>	<b>4.7±0.9</b>	<b>0.36±0.01</b>	<b>13.3±0.7</b>	<b>104±47</b>	<b>0.7±0.1</b>	<b>11±1.9</b>	<b>63.04±2.1</b>	<b>811±99</b>	<b>123±23</b>	<b>71±6</b>
$\sigma$		<b>20</b>	<b>2.8</b>	<b>1.1</b>	<b>0.018</b>	<b>0.90</b>	<b>59</b>	<b>0.18</b>	<b>2.36</b>	<b>2.60</b>	<b>124</b>	<b>29</b>	<b>7</b>
r [%]		<b>64</b>	<b>78</b>	<b>76</b>	<b>95</b>	<b>93</b>	<b>43</b>	<b>74</b>	<b>78</b>	<b>96</b>	<b>85</b>	<b>76</b>	<b>90</b>
60 min	OR6	67	10.0	3.2	0.32	14.3±2.5	159	1	7.67	69.06±0.16	784±48	169	77
	OR7	66	17.6	4.3	0.24	11.5±0.7	41	0.3	11.69	62.08±0.22	712±20	103	59
	OR8	87	19.8	5.9	0.30	15.6±2.4	80	0.8	6.72	68.44±0.19	819±34	74	68
	OR9	80	14.1	4.1	0.28	13.0±0.8	36	0.2	9.36	63.15±0.27	628±20	90	87

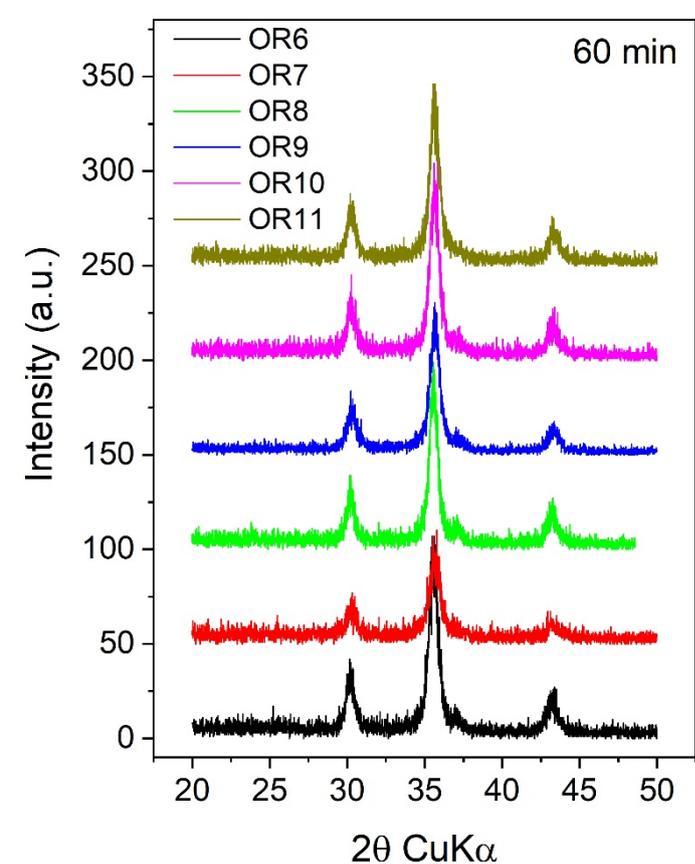
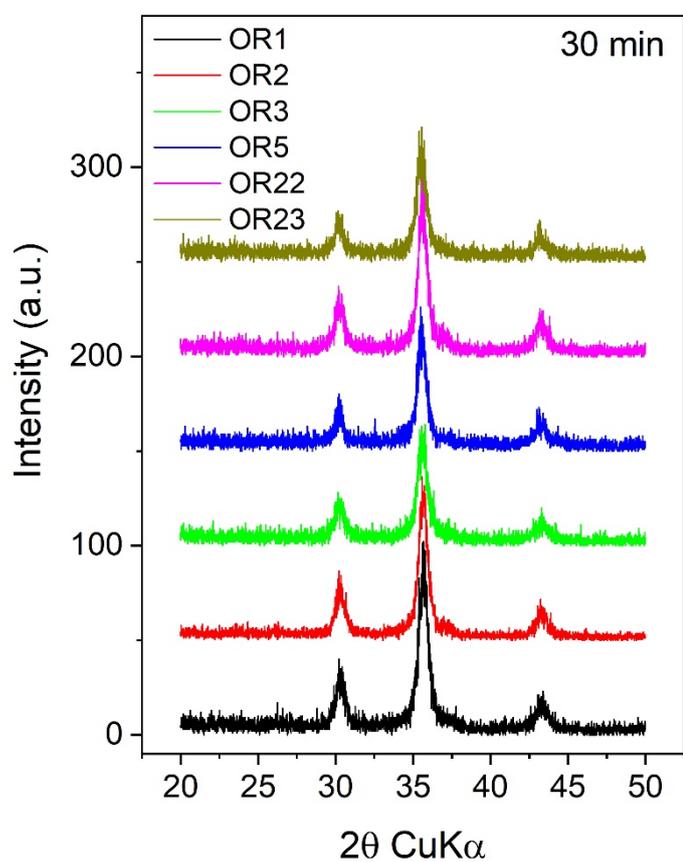
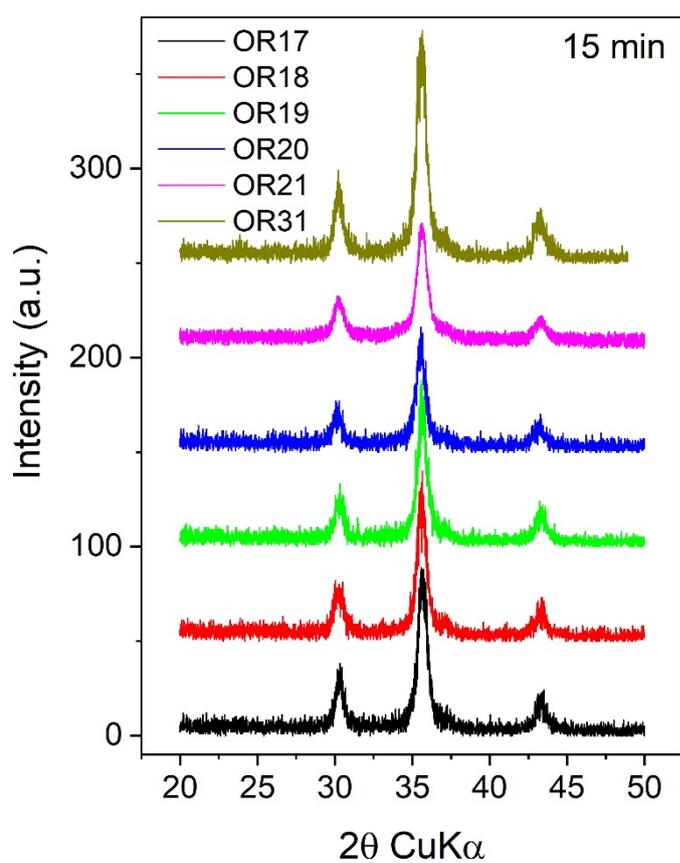
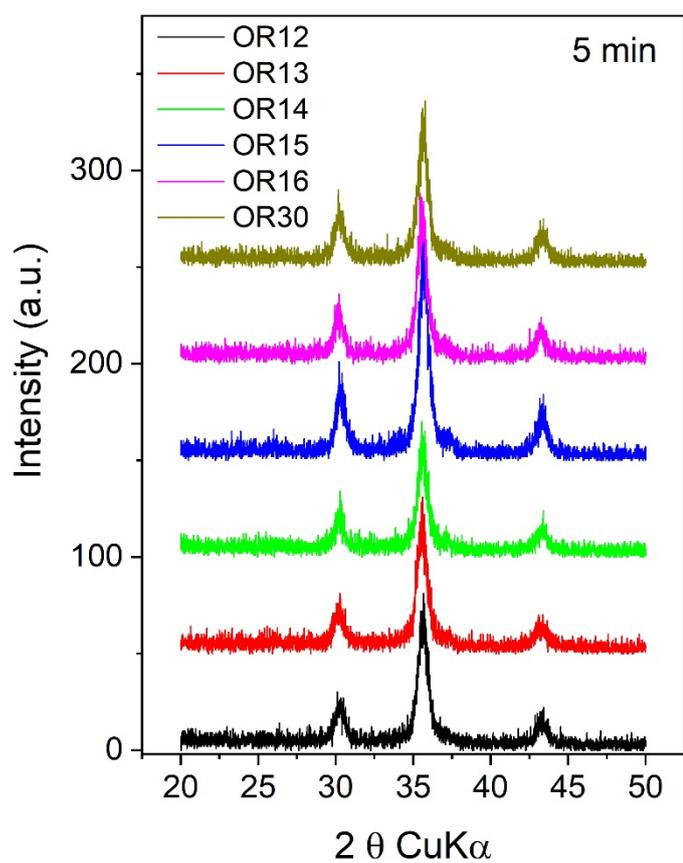
	OR1 0	51	22.0	4.4	0.20	13.7±1.6	192	1	8.56	66.32±0.31	902±47	186	106
	OR1 1	73	13.5	5.2	0.39	14.1±1.4	261	1	8.54	65.19±0.21	851±60	151	94
$\bar{x}$		<b>71±10</b>	<b>16±4</b>	<b>4.5±0.7</b>	<b>0.29±0.05</b>	<b>13.7±1.0</b>	<b>128±66</b>	<b>0.7±0.3</b>	<b>8.8±1.2</b>	<b>65.71±2.0</b>	<b>783±72</b>	<b>129±34</b>	<b>82±16</b>
$\sigma$		<b>12.5</b>	<b>4.5</b>	<b>0.9</b>	<b>0.066</b>	<b>1.26</b>	<b>83</b>	<b>0.34</b>	<b>1.55</b>	<b>2.55</b>	<b>90</b>	<b>42</b>	<b>15.8</b>
r [%]		<b>82</b>	<b>72</b>	<b>80</b>	<b>77</b>	<b>90</b>	<b>35</b>	<b>51</b>	<b>82</b>	<b>96</b>	<b>89</b>	<b>67</b>	<b>81</b>
120 min	OR2 4	59	18.2	4.4	0.24	14.0±0.5	616	0.8	12.79	69.18±0.12	1007±51	169	96
	OR2 5	43	19.0	5.7	0.30	13.6±0.9	282	0.8	12.48	65.71±0.10	887±45	137	66
	OR2 6	56	18.1	3.9	0.22	13.7±0.8	263	0.7	11.24	66.69±0.10	689±16	108	82
	OR2 7	74	18.1	4.2	0.23	13.6±0.3	207	0.7	10.41	69.01±0.25	869±9	108	57
	OR2 8	67	22.8	4.3	0.19	14.1±0.4	533	0.3	9.97	69.79±0.08	857±33	107	62
	OR2 9	66	21.1	3.7	0.18	12.0±1.3	284	0.7	11.08	63.53±0.01	884±44	96	63
$\bar{x}$		<b>61±8</b>	<b>20±1</b>	<b>4.4±0.5</b>	<b>0.23±0.03</b>	<b>13.5±0.6</b>	<b>364±121</b>	<b>0.7±0.1</b>	<b>11±0.9</b>	<b>67.3±1.8</b>	<b>866±74</b>	<b>121±20</b>	<b>71±11</b>
$\sigma$		<b>9.8</b>	<b>1.8</b>	<b>0.6</b>	<b>0.039</b>	<b>0.697</b>	<b>152</b>	<b>0.17</b>	<b>1.11</b>	<b>2.22</b>	<b>93</b>	<b>25</b>	<b>14</b>
r [%]		<b>84</b>	<b>91</b>	<b>86</b>	<b>83</b>	<b>95</b>	<b>58</b>	<b>76</b>	<b>90</b>	<b>97</b>	<b>89</b>	<b>79</b>	<b>80</b>

Table S2. Unscaled 1/20 samples for reference

	Exp.	Yield [%]	TEM			DRX Scherrer's size	PCS D <sub>SDV</sub> 0.1mgFe/mL		TG- ATD	Magnetic properties at room temperature		MAGNETIC HIPERTHERMIA 1mgFe/mL	
			D <sub>TEM</sub> [nm]	σ <sub>TEM</sub> [nm]	σ <sub>TEM</sub> /D <sub>TEM</sub>	D <sub>DRX</sub> [nm]	Z <sub>ave</sub> [nm]	PDI	Organic wt%	M <sub>s</sub> emu/g	γ emu/gT	286 kHz 18 kAm <sup>-1</sup>	92 kHz 50 kAm <sup>-1</sup>
													SAR [W/gFe]
30 min	ORD1	43.6	14.1	4.3	0.30	12±0.5	37	0.26	12.03	66.90±0.36	1120±68	98.2	71.7
	ORD2	23.3	13.3	3.1	0.23	12±0.9	40	0.34	12.20	67.06±0.16	945±44	69.8	42.2
	ORD3	19.7	15.0	3.8	0.26	13±1.4	44	0.37	21.21	60.04±0.11	863±54	98.1	52.7
	ORD4	18.5	14.1	3.8	0.27	12±0.7	646	0.49	9.66	68.21±0.11	952±59	111	42.1
	ORD5	19.8	13.9	3.8	0.28	12±1	41	0.32	13.52	65.92±0.06	870±41	94.3	47.5
	ORD6	21.4	13.8	4.0	0.29	13±0.9	42	0.32	12.21	68.03±0.18	1261±94	109	55.3
$\sigma$		<b>24.4±7</b>	<b>14.0±0.4</b>	<b>3.8±0.3</b>	<b>0.27±0.02</b>	<b>12.3±0.4</b>	<b>41±2</b>	<b>0.32±0.03</b>	<b>13.5±2.9</b>	<b>66.03±2.2</b>	<b>1002±114</b>	<b>96±10</b>	<b>52±8</b>
r [%]		<b>64</b>	<b>96</b>	<b>91</b>	<b>92</b>	<b>96</b>	<b>94</b>	<b>89</b>	<b>73</b>	<b>96</b>	<b>86</b>	<b>87</b>	<b>81</b>



**Figure S2** Measurements of the successive AMF heating and cooling cycles for selected samples showing the effects of the progressive destabilization of the colloids.



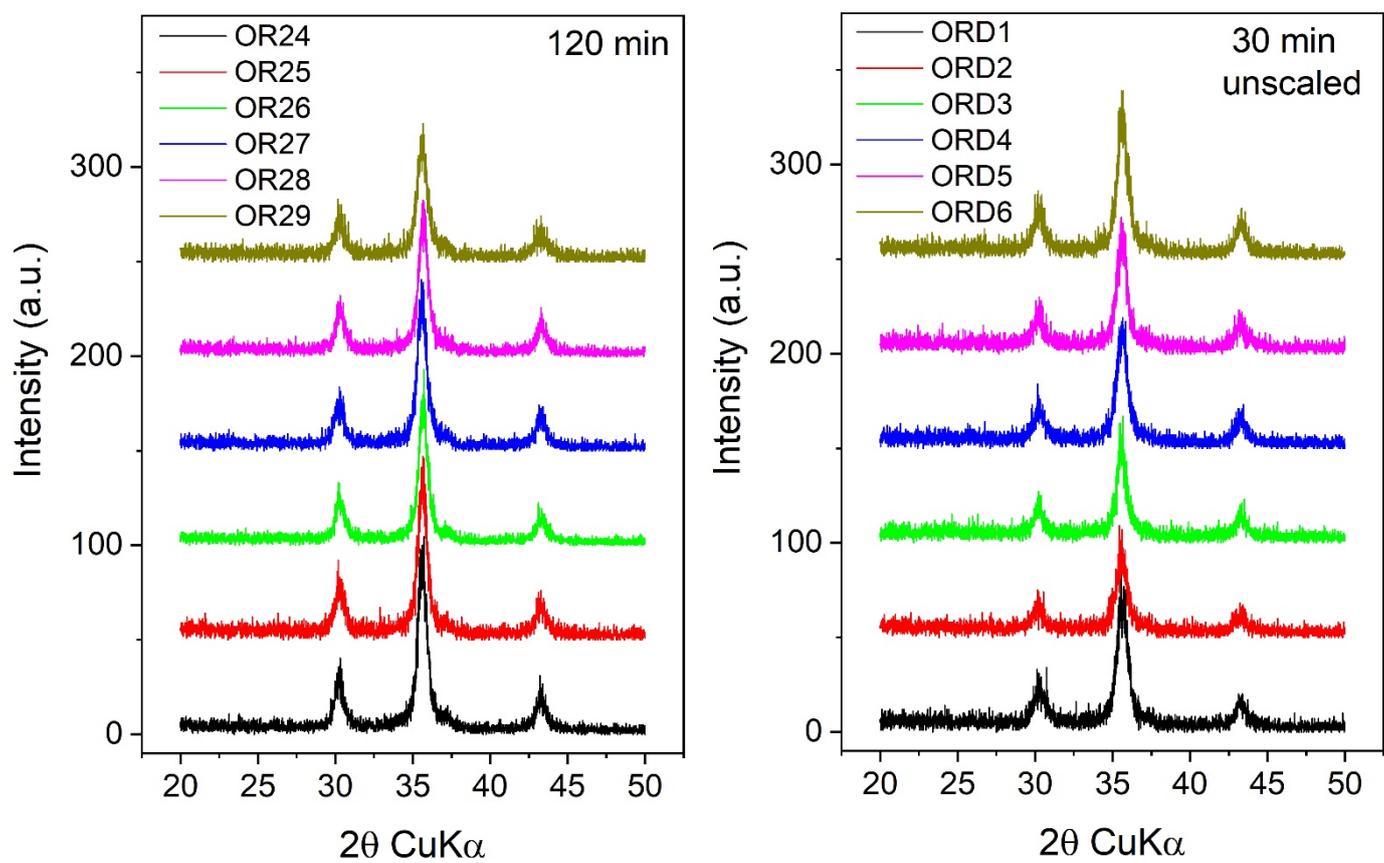
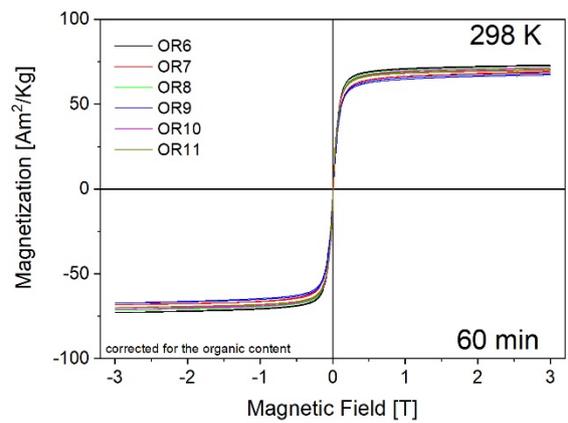
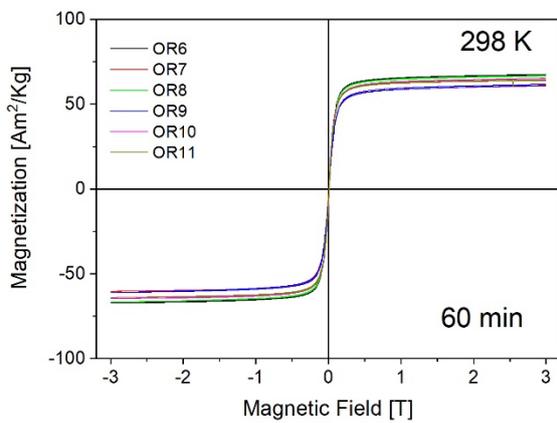
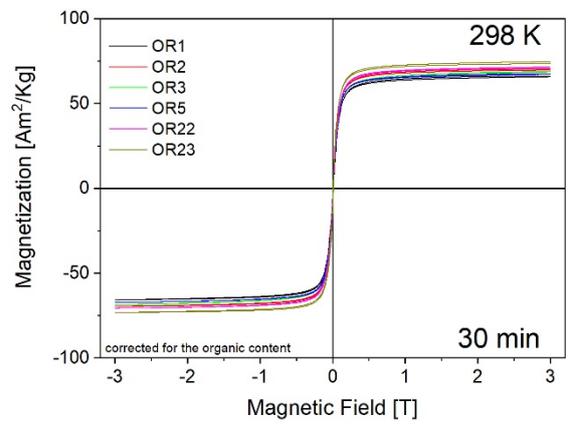
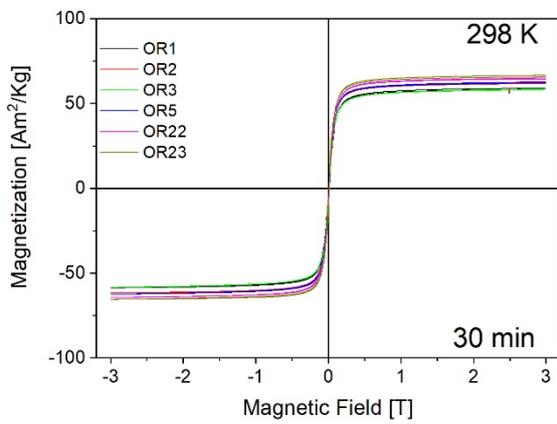
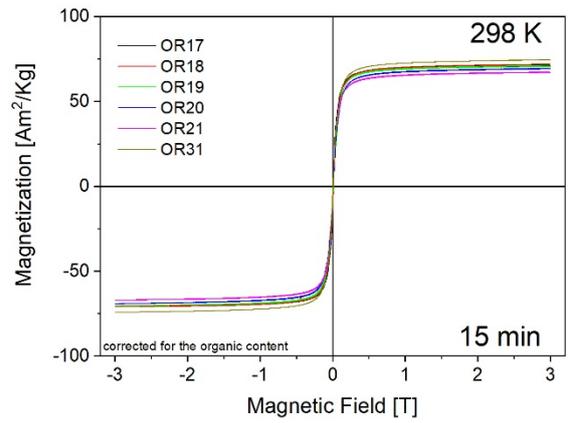
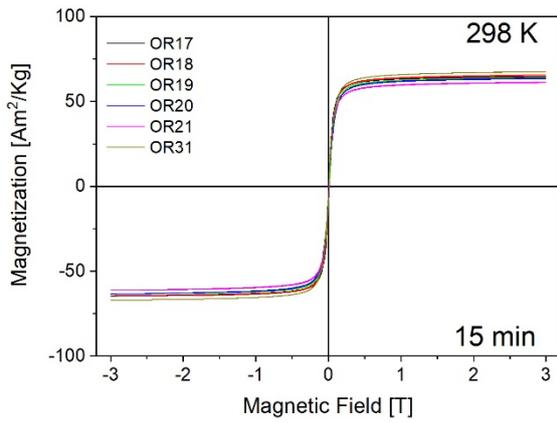
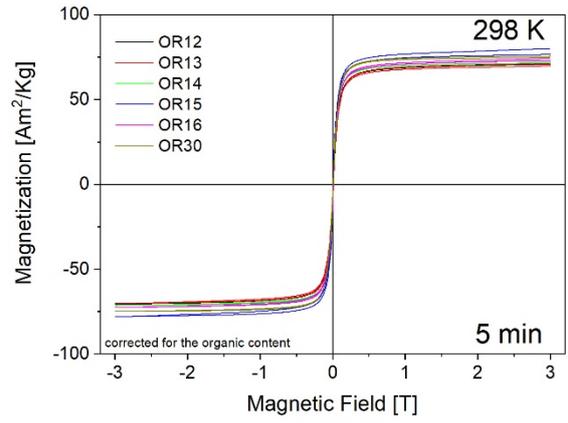
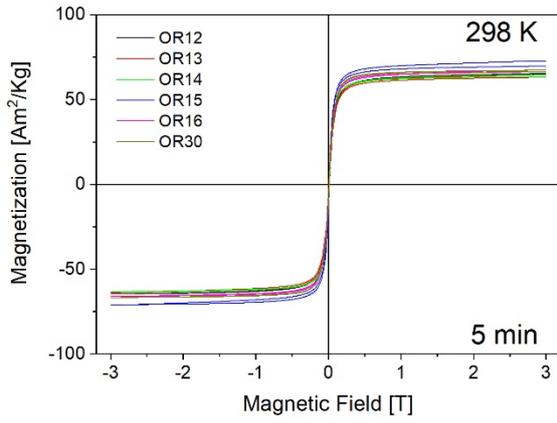
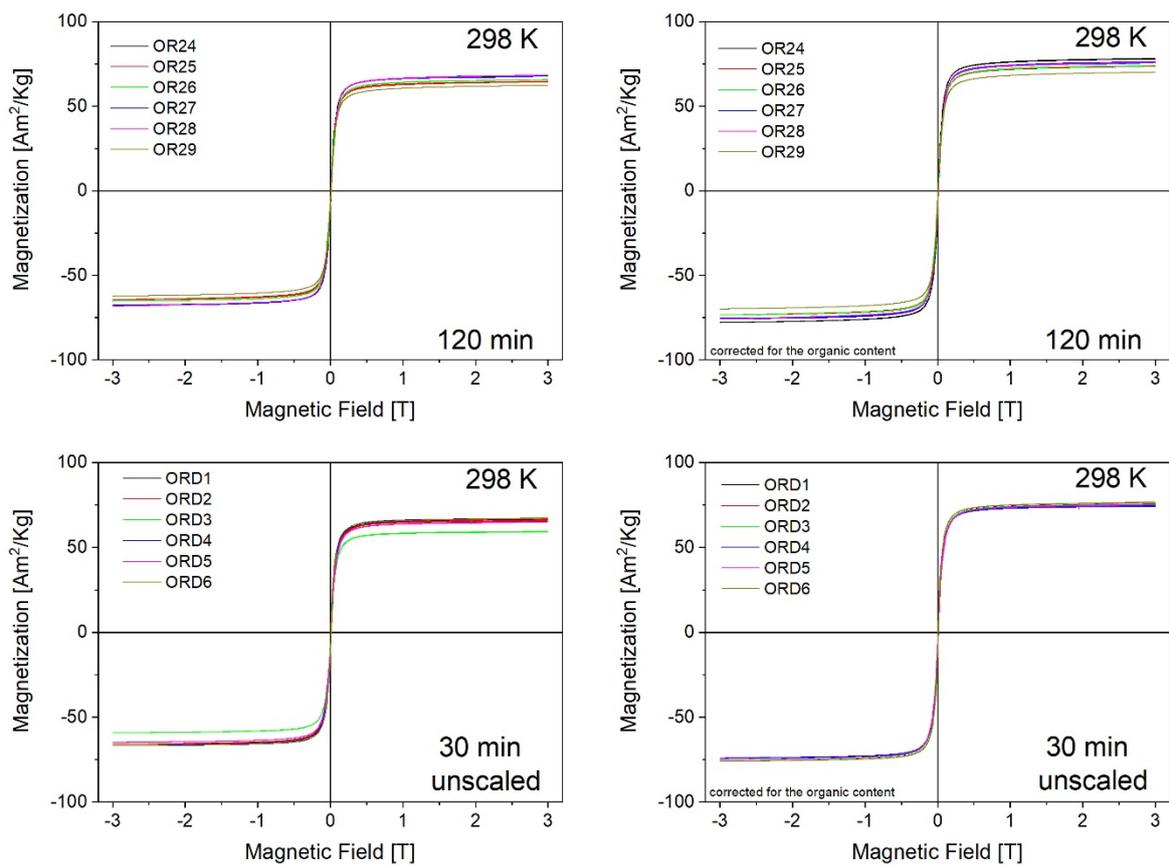
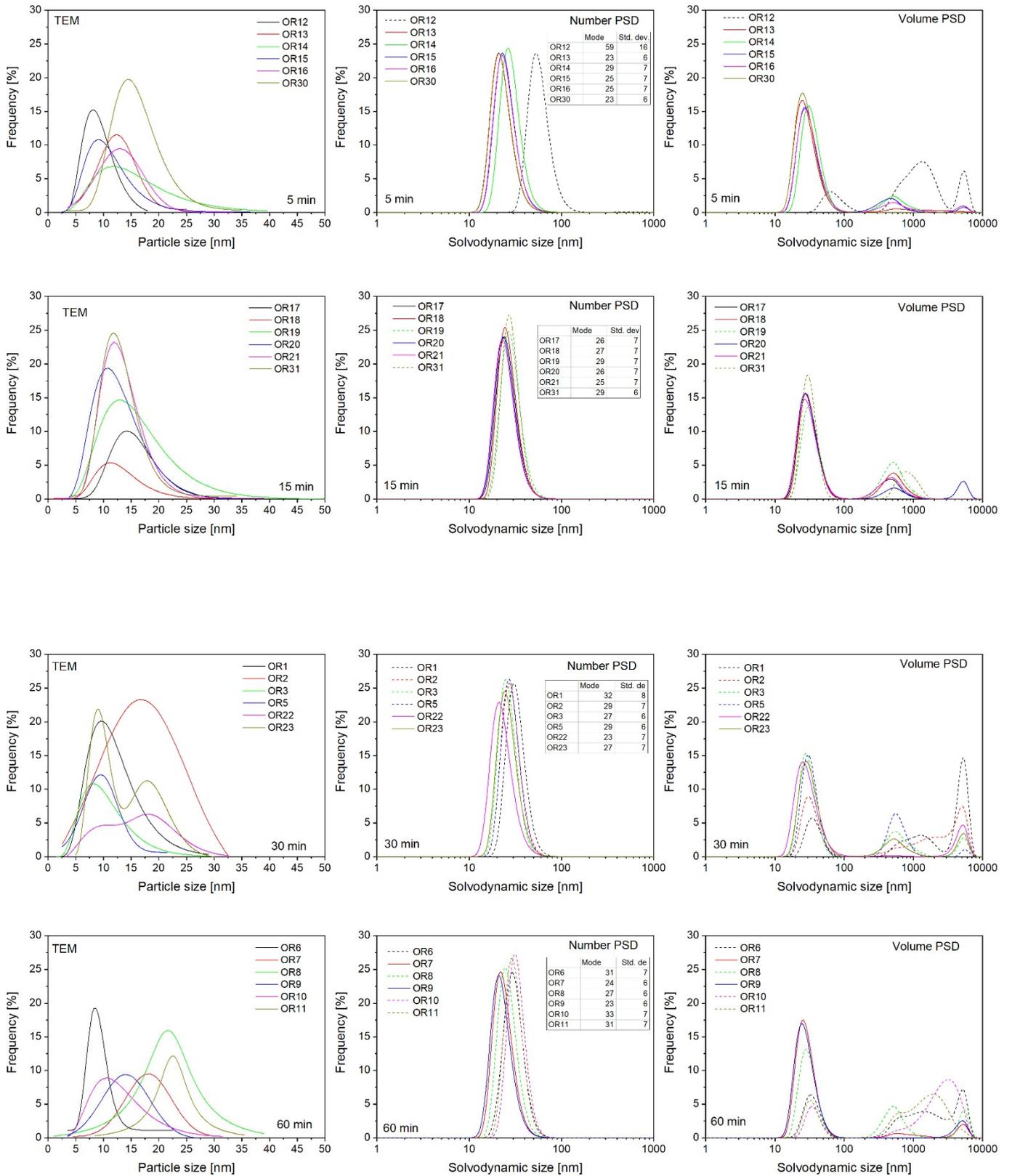


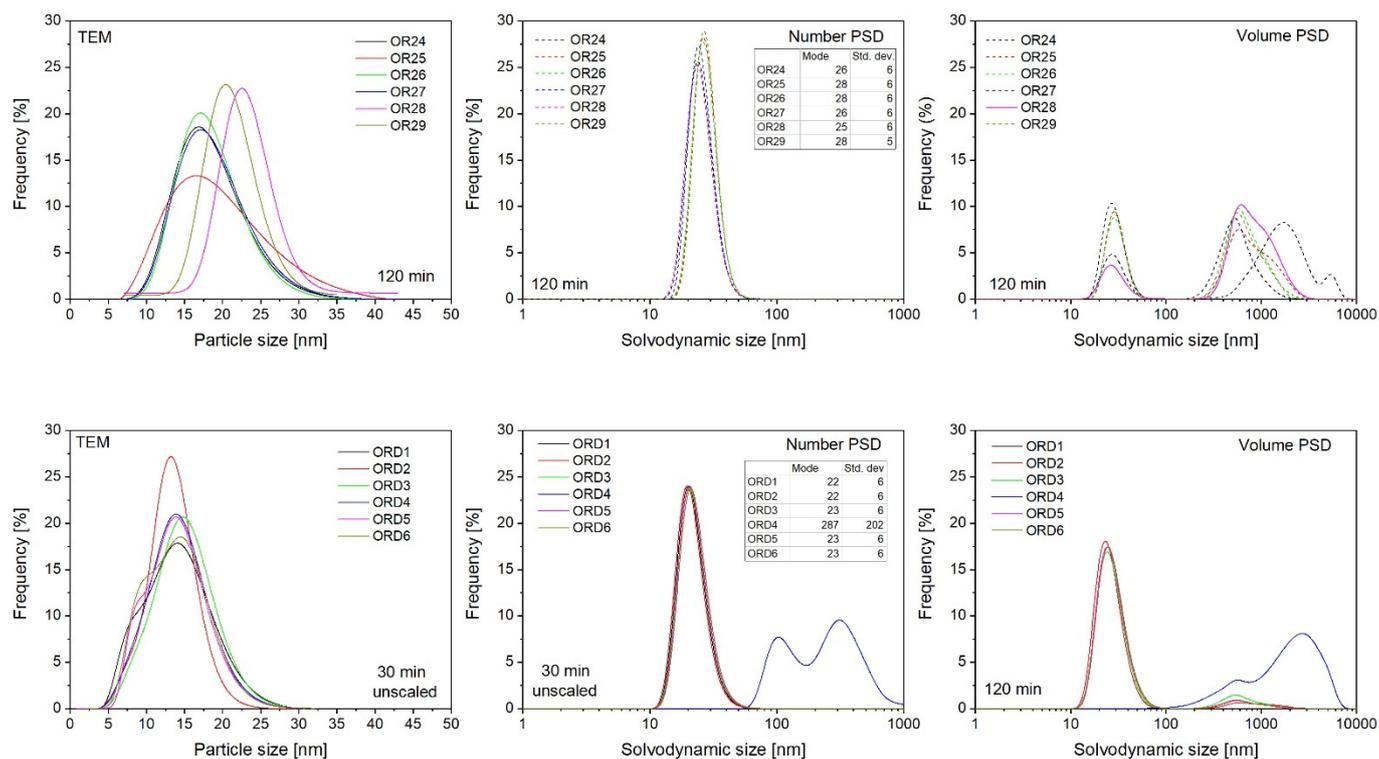
Figure S3: XRD diffraction patterns of all samples.



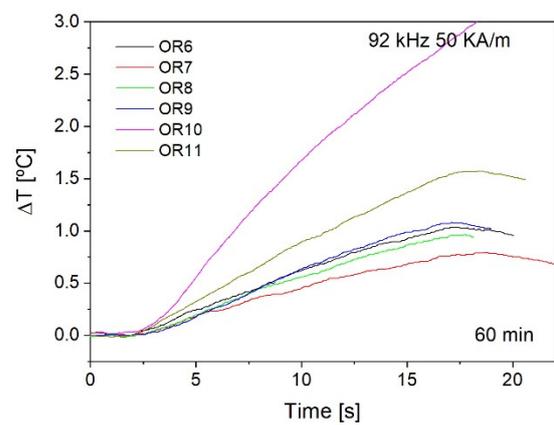
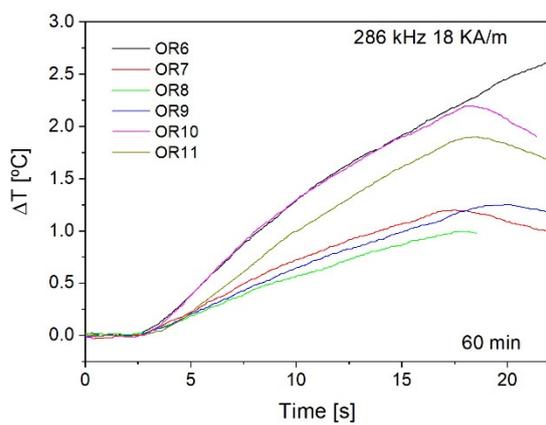
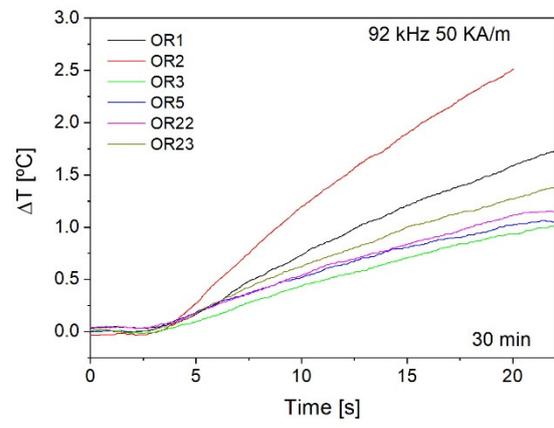
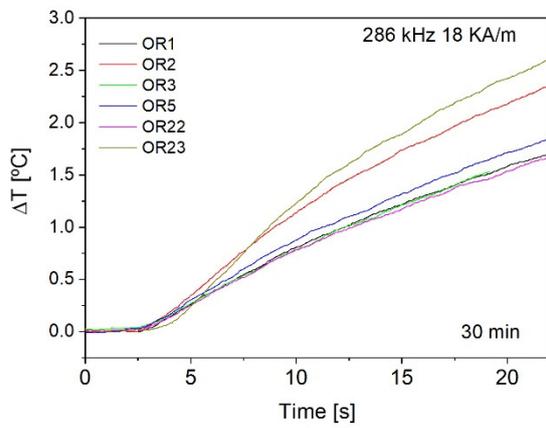
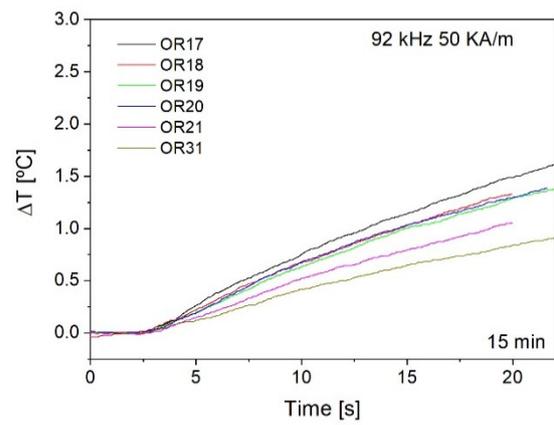
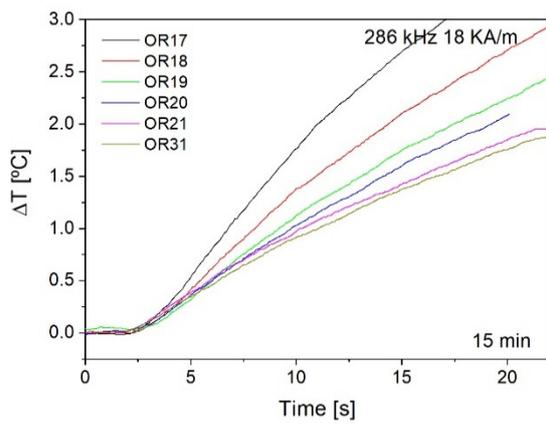
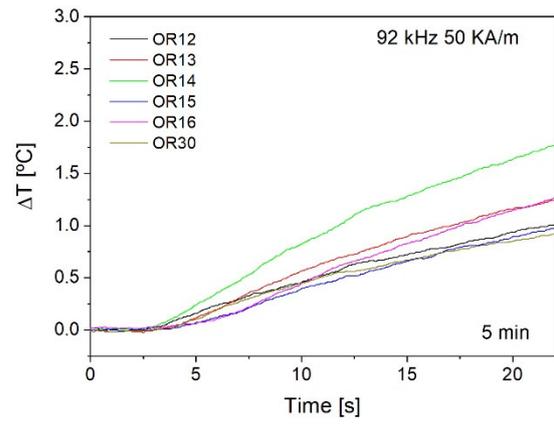
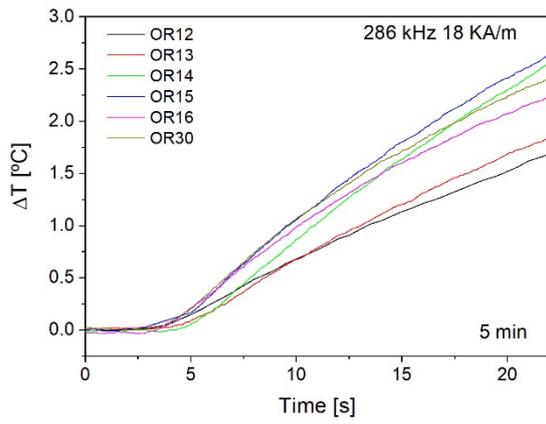


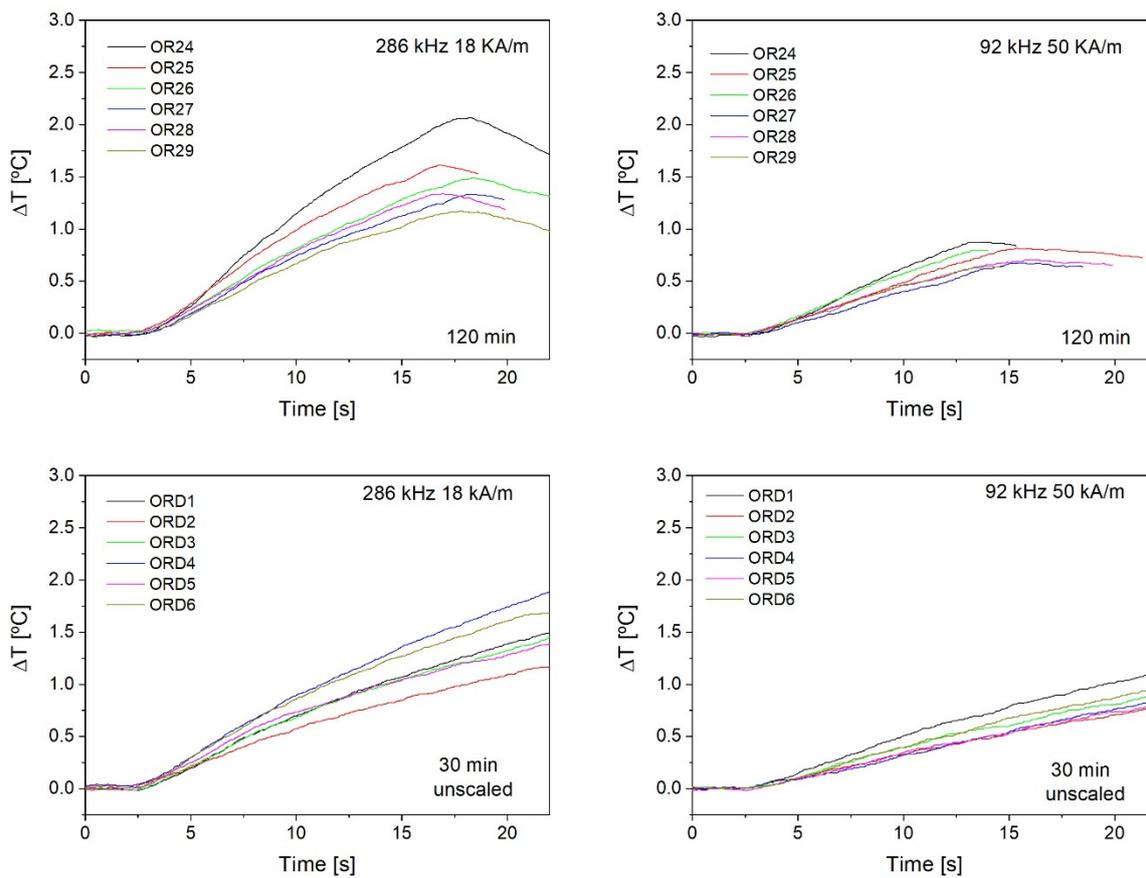
**Figure S4** Hysteresis cycles at room temperature of all the samples as measured (left) and corrected for the content of organic matter measured by TG-ATD (right).





**Figure S5** Particle size distributions of all the samples (left) and solvodynamic size distributions of their dispersions in toluene determined by PCS in number (center) and in volume (right). Polydisperse samples unable to be treated by the Contin algorithm are pictured with dash lines. The dispersion of the sample ORD4 was excluded from the statistical analysis.





**Figure S6:** Results of the magnetothermal experiments at 286 kHz 18 kA/m and 92 kHz 50 kA/m for all the samples. The samples were dispersed in oleic acid: toluene 1:7 v/v at 1 mg Fe/ml.