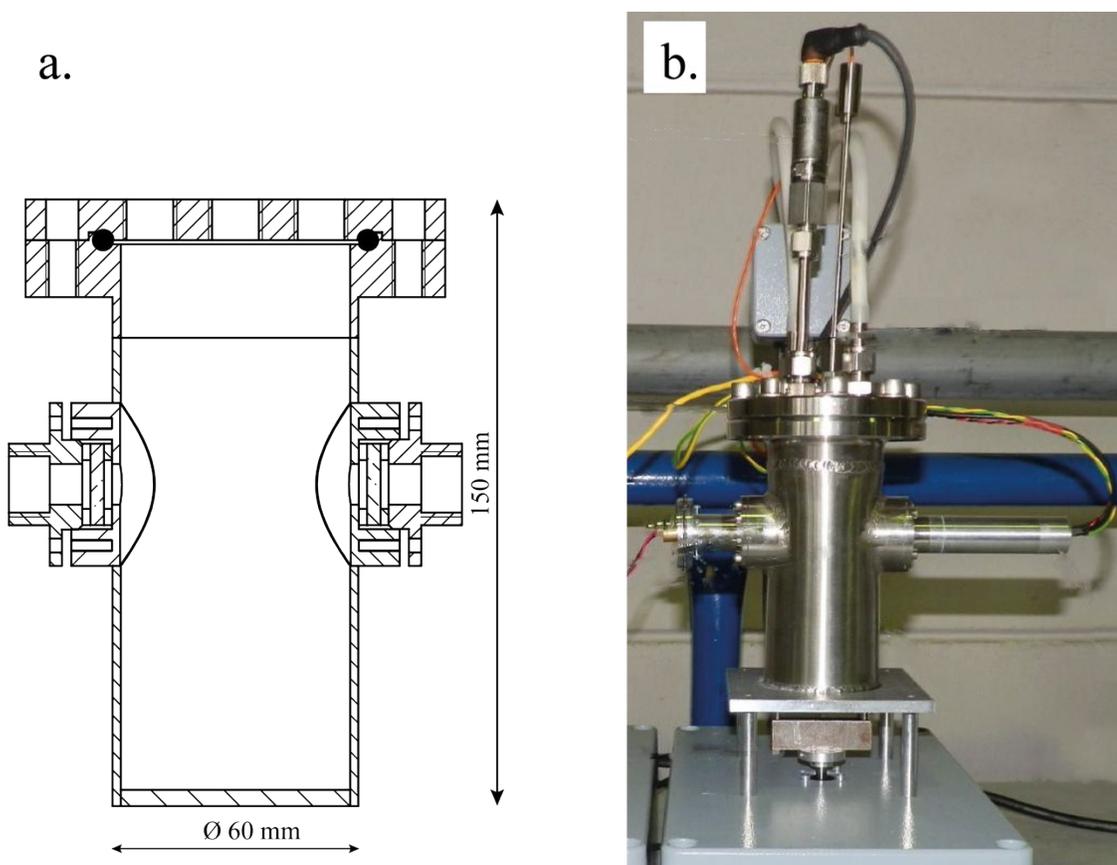
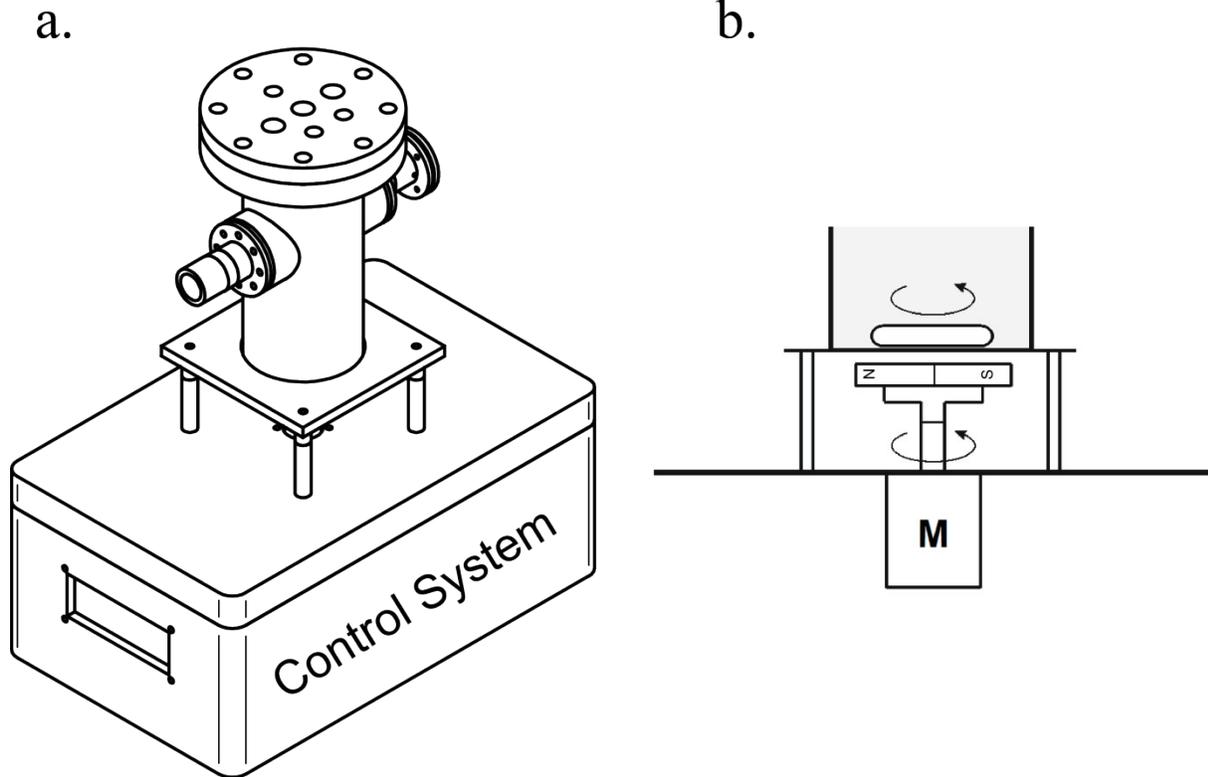


## Supplementary Information

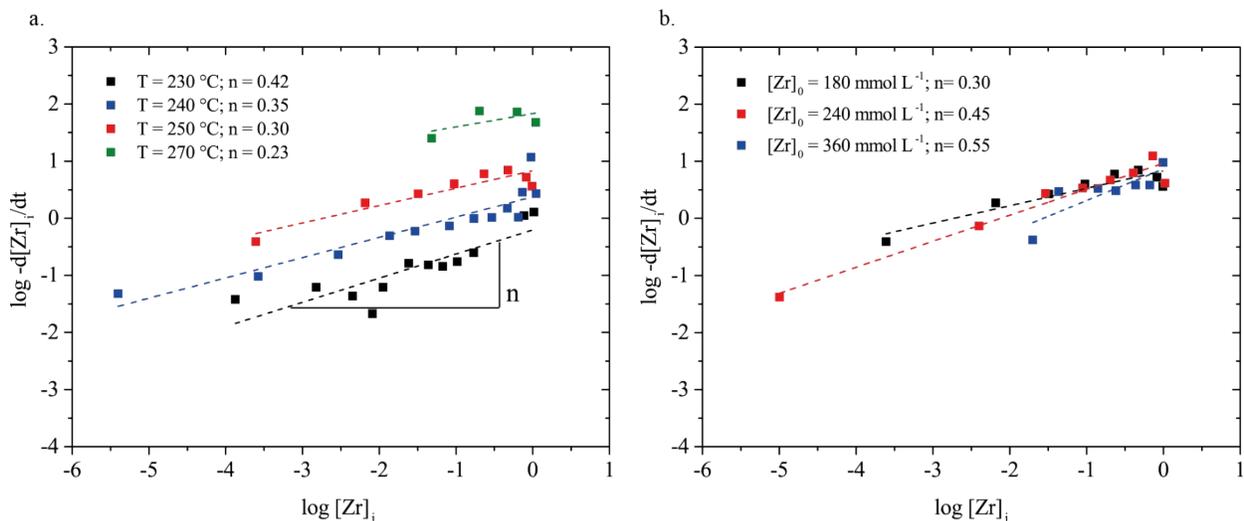
The 250 mL reactor system used for in situ transmission measurements



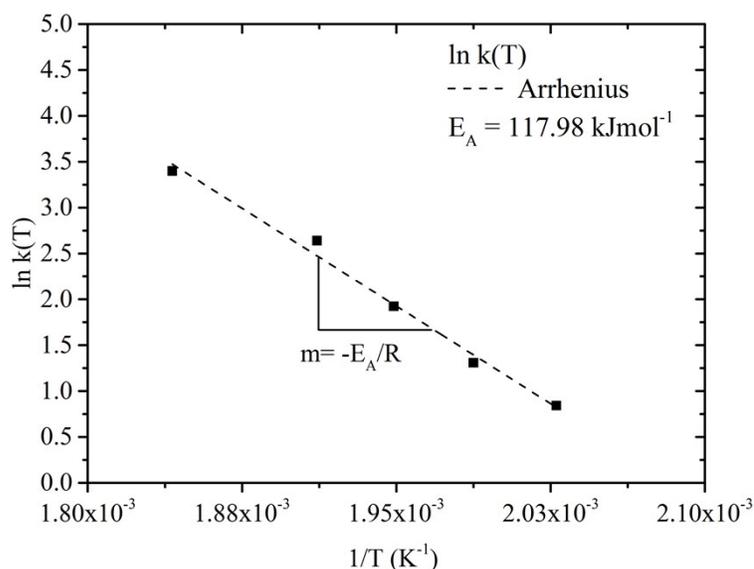
**Fig. S. 1** a. Illustration showing the reactor vessel and its dimensions; b. Reactor setup with the laser source (right) and detector (left) mounted to the borosilicate windows. Heat cartridges as well as the pressure and temperature sensors are located at the top part of the reactor.



**Fig. S. 2** a. Illustration of the reactor vessel positioned on top of the control and agitation system. The control system is connected to a personal computer which allowed full control and data acquisition via a LabVIEW program, b. Agitation was realized via a self-build magnetic stirrer.



**Fig. S. 3** Differential method applied to determine the reaction order of the nonaqueous sol-gel synthesis for different a. process temperatures with an initial precursor concentration  $[\text{Zr}]_0$  of  $180\text{ mmol L}^{-1}$  and b. initial precursor concentrations at  $250\text{ }^\circ\text{C}$ .

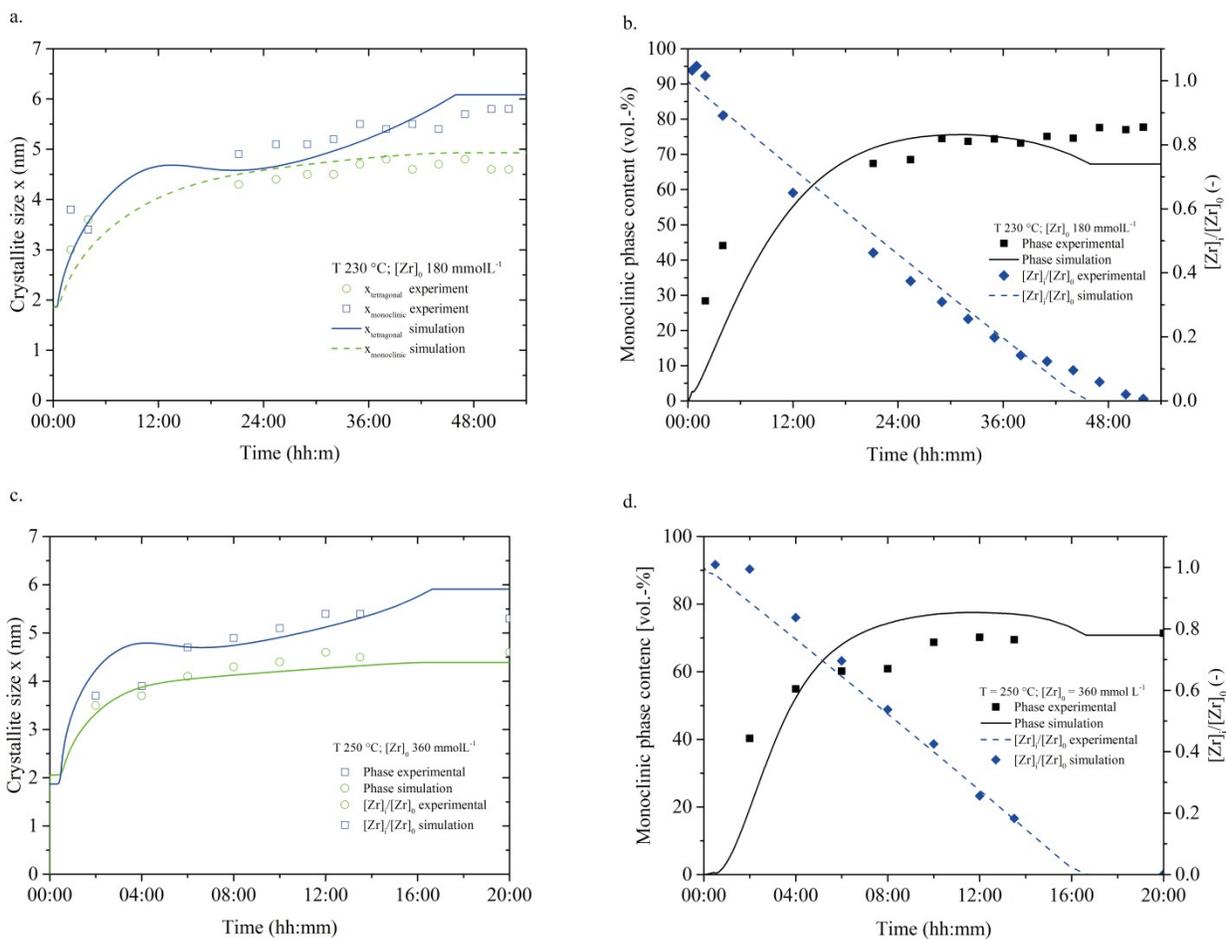


**Fig. S. 4.** Arrhenius plot illustrating the temperature influence on the chemical reaction rate with an activation energy value  $E_A$  determined to  $117.98 \text{ kJ mol}^{-1}$ .

**Table S. 1**

Nanoparticles synthesized at different process conditions T (°C); [Zr] <sub>0</sub> (mmolL <sup>-1</sup> )	Tetragonal crystallite size $x_t$ (nm)	Monoclinic crystallite size $x_m$ (nm)	Monoclinic phase content (vol.-%)	Number of monoclinic particles to total number of particles $N_{\text{mono}}/N_{\text{total}}$ (-)
T = 220; [Zr] <sub>0</sub> = 180	4.9	4.3	60.6	0.68
T = 270; [Zr] <sub>0</sub> = 180	3.6	2.7	18.2	0.34
T = 250; [Zr] <sub>0</sub> = 180	5.9	5.0	81.5	0.87
T = 230; [Zr] <sub>0</sub> = 180	5.7	4.6	74.6	0.84
T = 240; [Zr] <sub>0</sub> = 180	6.1	4.7	82.0	0.90
T = 220; [Zr] <sub>0</sub> = 360	6.6	5.8	81.5	0.87
T = 230; [Zr] <sub>0</sub> = 360	6.2	4.2	75.3	0.90
T = 250; [Zr] <sub>0</sub> = 360	5.4	4.9	63.0	0.69
T = 270; [Zr] <sub>0</sub> = 360	4.1	3.7	31.4	0.46
T = 220; [Zr] <sub>0</sub> = 240	4.9	4.4	62.0	0.69
T = 230; [Zr] <sub>0</sub> = 240	6.1	4.3	81.5	0.90
T = 240; [Zr] <sub>0</sub> = 360	7.0	6.5	84.3	0.92
T = 250; [Zr] <sub>0</sub> = 240	5.3	4.6	56.9	0.66
T = 270; [Zr] <sub>0</sub> = 240	3.9	2.5	25.9	0.57
T = 250; [Zr] <sub>0</sub> = 180*	3.9	3.1	22.2	0.36

\*Initial reaction mixture contained ZrO<sub>2</sub>-nanoparticle seeds.



**Fig. S. 5 a. & b.** Validation of experimental and simulation results for a synthesis carried out at 230 °C with an initial precursor concentration of 180 mmol L<sup>-1</sup>; c. & b. Validation of experimental and simulation results for a synthesis carried out at 250 °C with an initial precursor concentration of 360 mmol L<sup>-1</sup>.