Supporting Information

Continuous Microfluidic Synthesis of Pd Nanocubes and PdPt Core-Shell Nanoparticles and Their Catalysis of NO_2 Reduction

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S1. Microfluidic synthesis optimization of Pd nanocubes

For the development of the Pd nanocube synthesis an investigation of reaction parameters and their influence on the particle size and morphology was performed to find optimum conditions to produce uniform Pd nanocubes. Figure S1 shows TEM images of Pd nanoparticles synthesised with variation of residence time, CTAB concentration and temperature. Table S1 lists experimental details of reaction conditions for respective particle batch from figure S1.

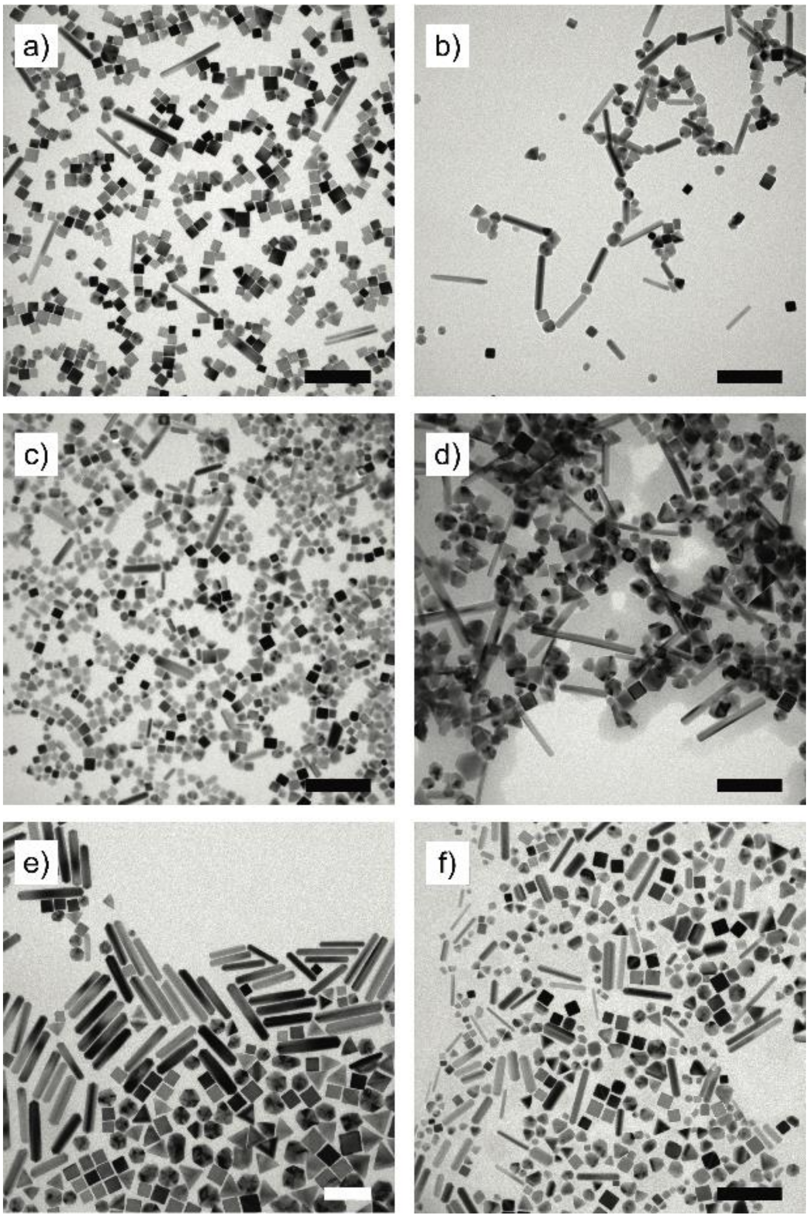

Figure S1. TEM images of Pd nanocubes synthesized under different reaction conditions. The residence time is increased to a) 10 min and b) 20 min. Concentration of CTAB is in c) 4.17 mM and d) 37.5 mM. Variation of reaction temperature in e) 60°C and f) 130°C. Scale bars are 100 nm.

Table S1. Experimental details for the microfluidic synthesized Palladium nanoparticles shown in Figure S1. Reaction parameters evaluated are residence time, CTAB concentration and temperature.

Nanoparticle batch	Residence time (min)	Concentration CTAB (mM)	Temperature (°C)
Standard conditions	3	26.1	96
Fig S1a)	10	26.1	96
Fig S1b)	20	26.1	96
Fig S1c)	10	8.7	96
Fig S1d)	10	78.4	96
Fig S1e)	3	26.1	60
Fig S1f)	3	26.1	130

S2. Millifluidic synthesis of Pd nanocubes

The synthesis of cubic Pd nanoparticles was successfully upscaled to a milliliter sized reactor while still retaining morphology and monodispersity of the particles, shown in the TEM image and histogram of particle size distribution in Figure S2.

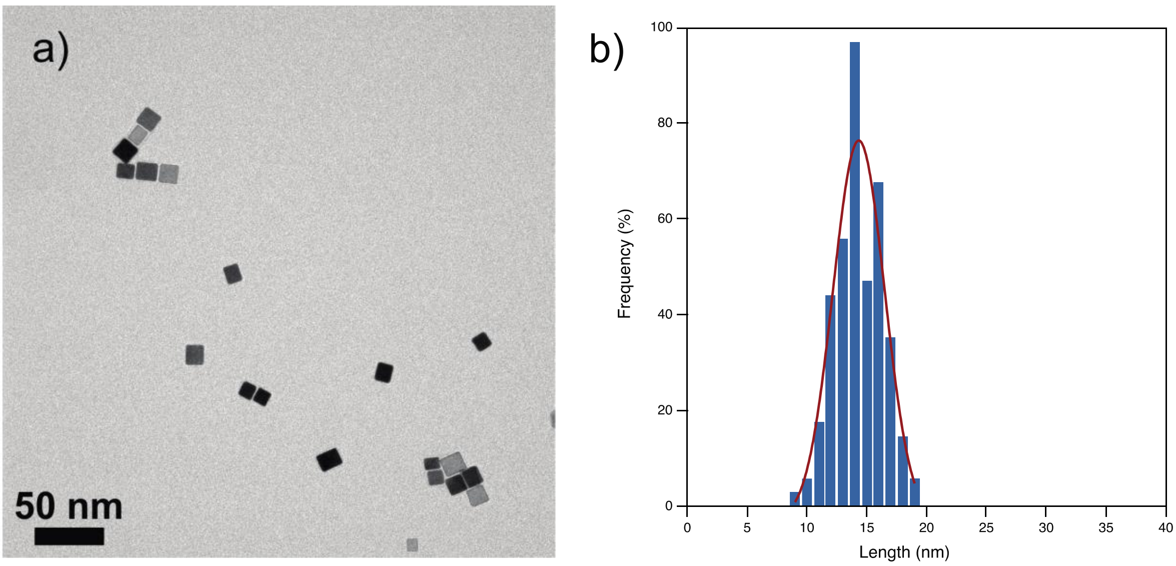

Figure S2. a) TEM image of Pd nanocubes produced in a millifluidic reactor and b) histogram of particle size distribution and Gaussian curve fit to the data. Pd nanocubes have an average size of 14 nm ± 11 %.

S3. Particle yield of Pd nanocubes

Table S2 lists the particle yield measured by MP-AES, of synthesized Pd nanocubes from the batch, microfluidic and millifluidic reactor.

Table S2. Presentation of the Pd yield and the from the synthesis of Pd nanocubes in batch, microfluidic and millifluidic reactors. The loss represents the difference in amount Pd precursor added to the reaction and the amount measured after reaction.

Reactor	Yield (%)	Loss (%)
Batch	63	3
Microfluidic	94	0
Millifluidic	33	35

S4. Microfluidic synthesis optimization of PdPt nanoparticles

In this work bimetallic core-shell PdPt nanoparticles with varying molar ratios of the metals were synthesized using a microfluidic reactor. The molar ratio of Pd:Pt (6:1, 3:1, 1:1) in the nanoparticles is varied by mixing different amount of H_2PdCl_4 and H_2Cl_6Pt solution in the metal precursor solution stream. The influence of reaction temperature on the morphology of formed PdPt (1:1) core-shell nanoparticles are investigated by synthesizing particles at 60°C and 130°C, and TEM images of the resulting particles as shown in Figure S3.

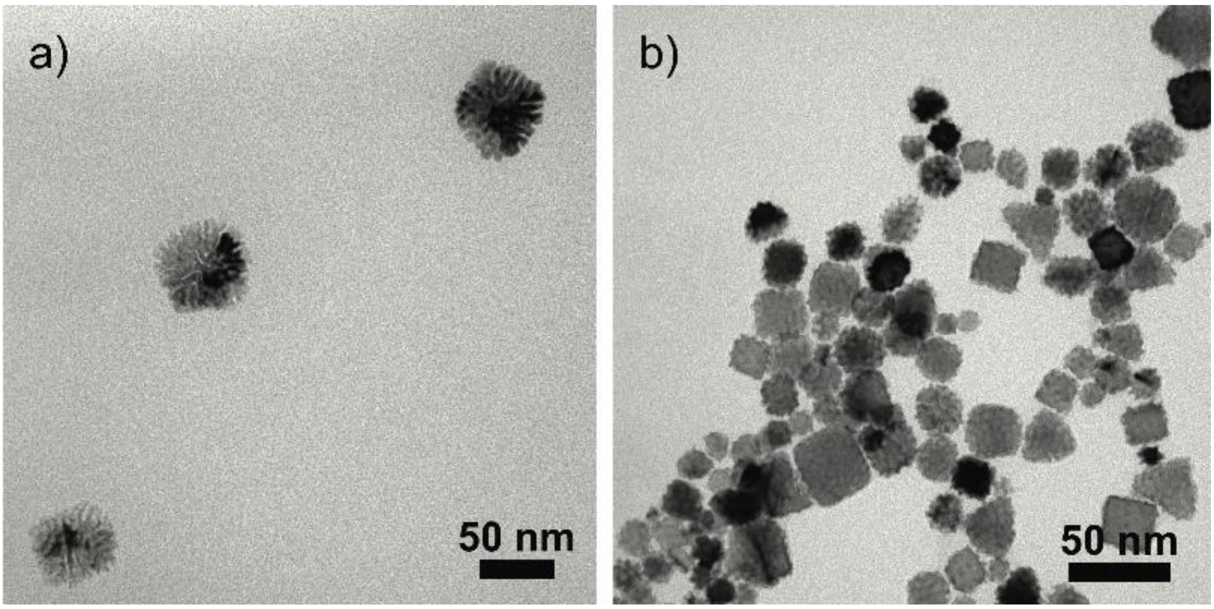

Figure S3. TEM images of PdPt (1:1) core-shell nanoparticles synthesized in different temperatures; a) 60°C and b) 130°C.

S5. Morphological evaluation of Pd nanocubes and PdPt core-shell nanoparticles after catalytic model reaction

Analysis of the shape and size of Pd nanoparticles and core-shell PdPt (1:1) nanoparticles after treatment in reaction conditions at elevated temperatures is evaluated by Scanning Electron Microscopy (SEM), and is presented in Figure S4.

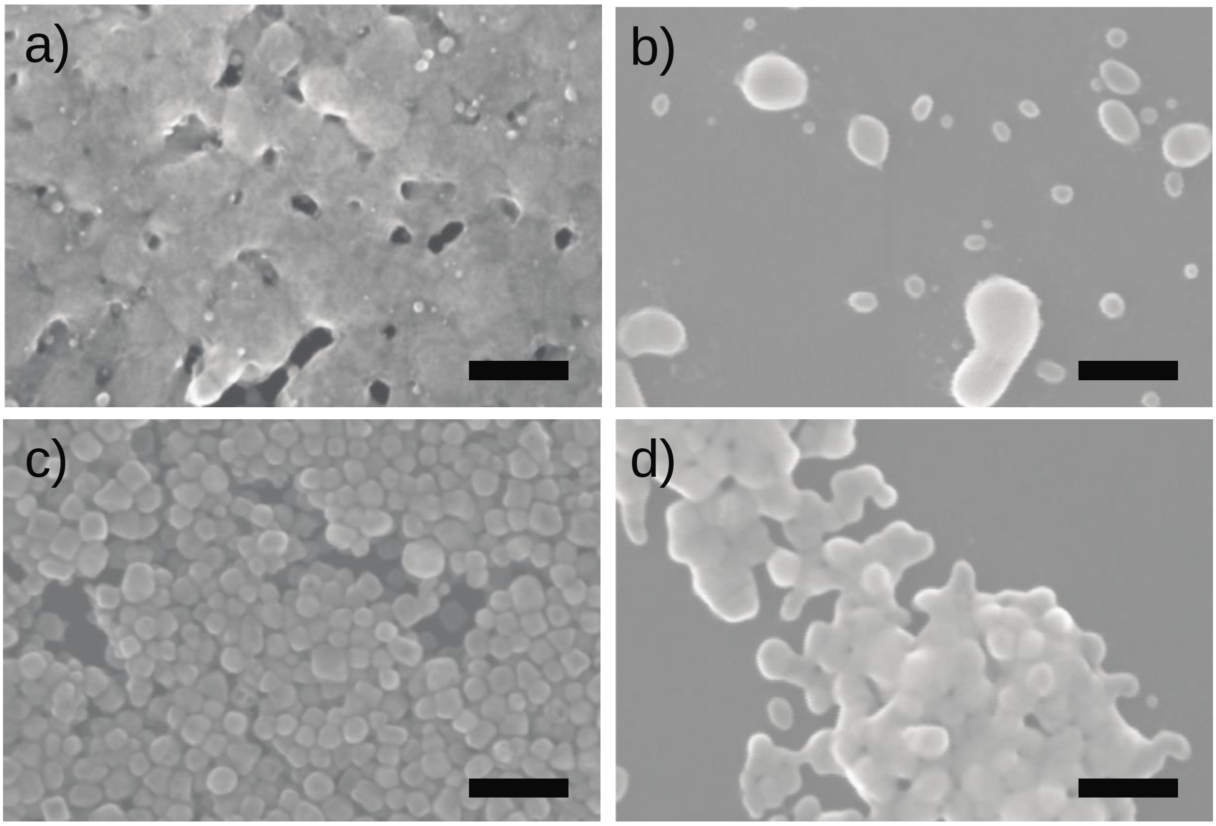

Figure S4. SEM images of a) Pd nanocubes and c) PdPt (1:1) nanoparticles deposited on Si substrates after treatment in 50-220°C temperature interval in 2200 ppm NO_2, 2.2 % H_2 in $Ar_{(g)}$. b) Pd nanocubes and d) PdPt (1:1) core-shell nanoparticles after treatment in 50-390°C temperature interval in 2200 ppm NO_2, 2.2 % H_2 in $Ar_{(g)}$. Scale bars are 100 nm.

S6. Direct H_2-deNO_2 activity of Pd nanocubes and PdPt core-shell nanoparticles

To evaluate the catalytic activity of the nanoparticles a temperature programmed NO_2-reduction experiment was performed. To exclude effects coming from the reactor, a blank experiment with an empty reactor was performed, see Figure S5.

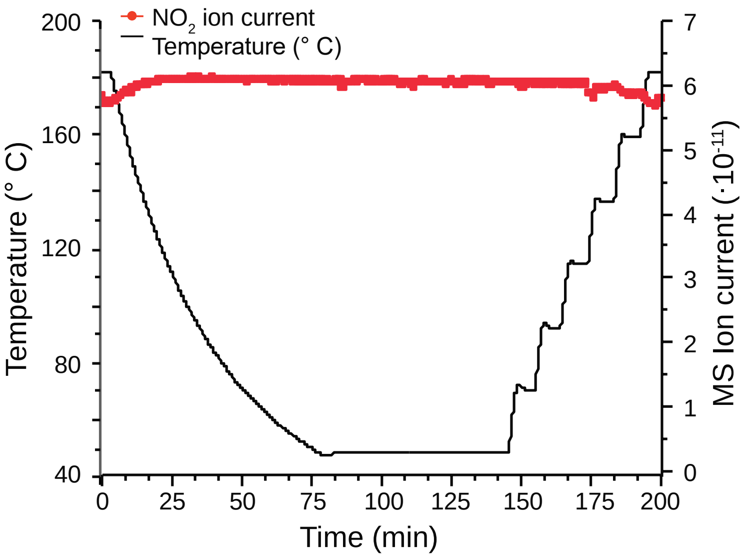

Figure S5. Temperature-dependent NO_2 mass spectrometer readings for an empty reactor. The reaction takes place in a mixture of 2200 ppm NO_2 and 2.22 % H_2 in Ar carrier gas , supplied at a 6 ml/min flow rate. Before temperature ramp up, the reactor was exposed to the reaction mixture at 48 ^{o}C for 3 hours