Supplementary Information

Synergizing ICP-MS, STEM-EDXS, and SMPS single particle analytics exemplified by superlattice L1_0 Pt/Fe aerosol nanoparticles produced by spark ablation

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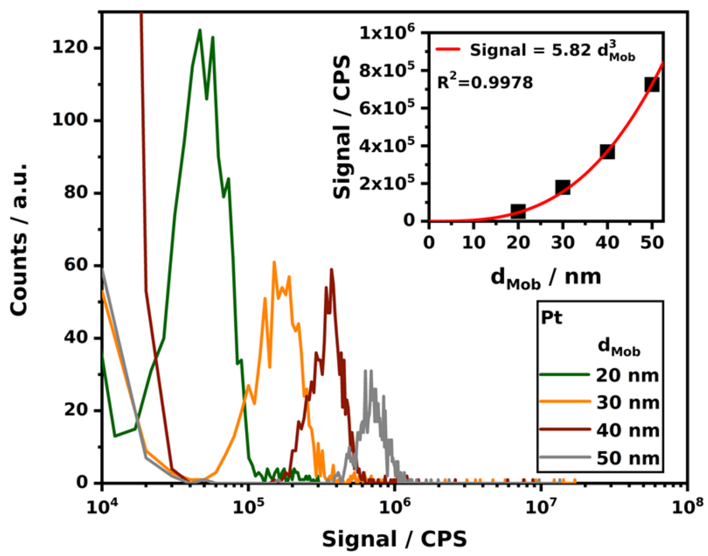

Fig. S1: Calibration of the spICP-MS device with spherical Pt NPs. The inset shows the derived mean signal response over the particle diameter.

Fig. S1 shows the raw data (signal distributions) of the spICP-MS, which was fed with spherical Pt aerosol NPs, classified at 20, 30, 40, and 50 nm by a DMA for a device calibration. From the mean signal response, a fit can be drawn to correlate the spICP-MS signal to a mobility-equivalent particle size (inset).

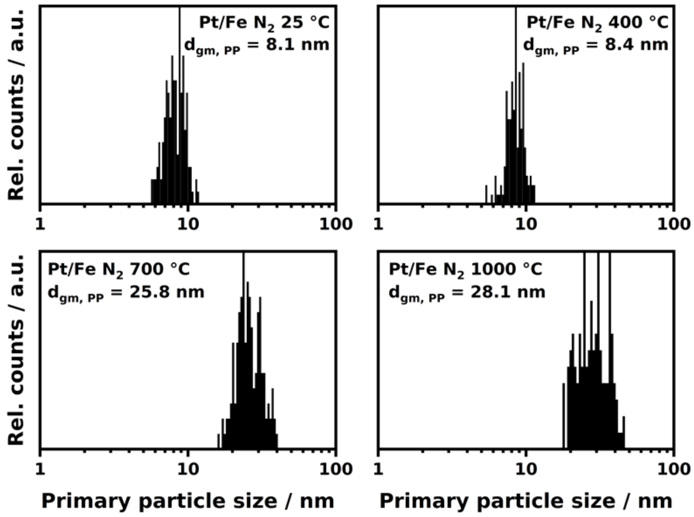

Fig. S2: Primary particle size distributions for subsequently progressing sintering of Pt/Fe agglomerates under N_2.

Fig. S2 shows the primary particle size distributions of Pt/Fe agglomerate particles, which were sintered under N_2. Interestingly, the TEM imaging revealed a larger primary particle size for Pt/Fe_2O_3 NPs sintered at 700 °C compared to Pt/Fe NPs sintered under H_2 at the same temperature (see Fig. S6). It can be hypothesized that characteristic coalescence times may change for particles made of Pt/Fe_2O_3, Pt/Fe in fcc, or Pt/Fe in $L1_0$ crystal configuration, which in turn may influence the primary particle structure at a certain temperature.

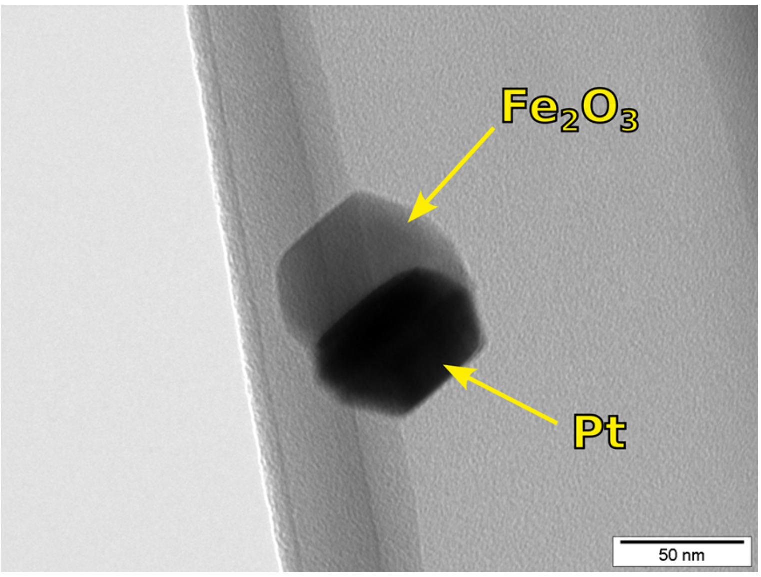

Fig. S3: TEM micrograph of Pt/Fe_2O_3 NPs sintered at 1450 °C.

Fig. S3 shows the janus-like particle morphology of Pt/Fe_2O_3 NPs after sintering at 1450 °C. The darker material can be assigned to Pt, and the brighter one to Fe_2O_3. Both segregated materials reached the thermodynamically favorable and compact sphere-like shape.

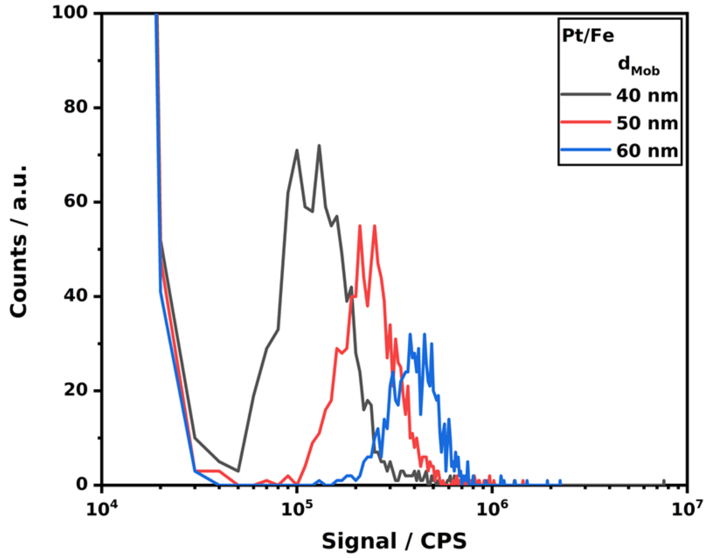

Fig. S4: spICP-MS signal response for Pt/Fe NPs.

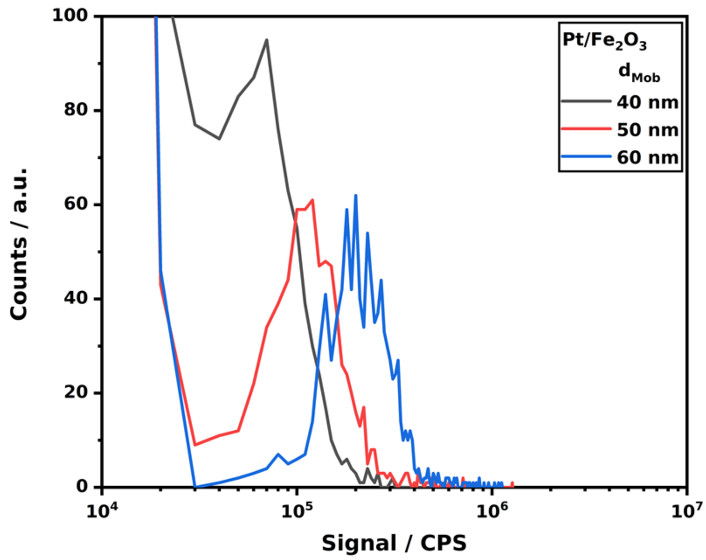

Fig. S5: spICP-MS signal response for Pt/Fe_2O_3 NPs.

Fig. S4 and S5 show the raw data (signal distributions) for alloyed and segregated NPs, respectively. From the signal distributions shown, the composition of single particles can be calculated.

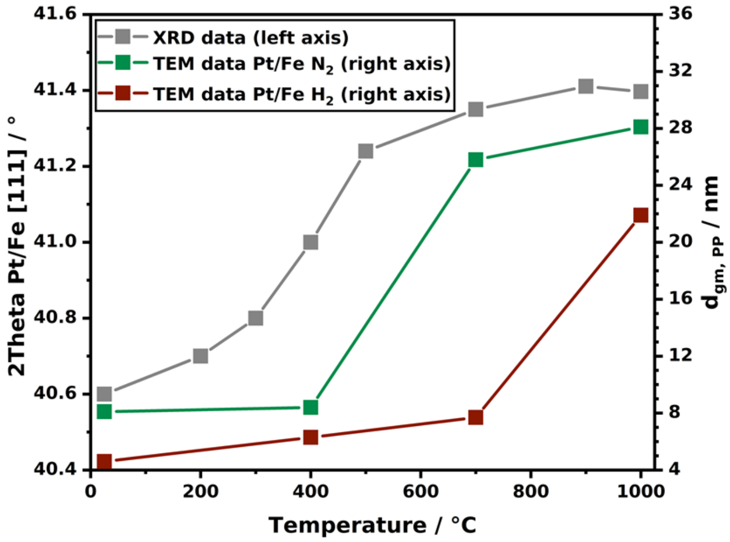

Fig. S6: Peak shift of the Pt/Fe [111] reflex with increasing sintering temperature and primary particle geometric mean diameters of Pt/Fe NPs sintered under N_2 or H_2, according to Fig. 3 and Fig. S2.

Fig. S6 shows the temperature-resolved peak shift of the Pt/Fe [111] reflex, indicating the formation of Pt/Fe in $L1_0$ order, as reported by Yu et al.[1] The peak shift implies the $L1_0$ formation to occur at around 400-600 °C.

References

1 J. Yu, W. Gao, F. Liu, Y. Ju, F. Zhao, Z. Yang, X. Chu, S. Che and Y. Hou, *Sci. China Mater.*, 2018, **61**, 961–968.