Supplementary Materials: Structural Changes of Highly Active Pd/MeO$_x$ (Me = Fe, Co, Ni) during Catalytic Methane Combustion

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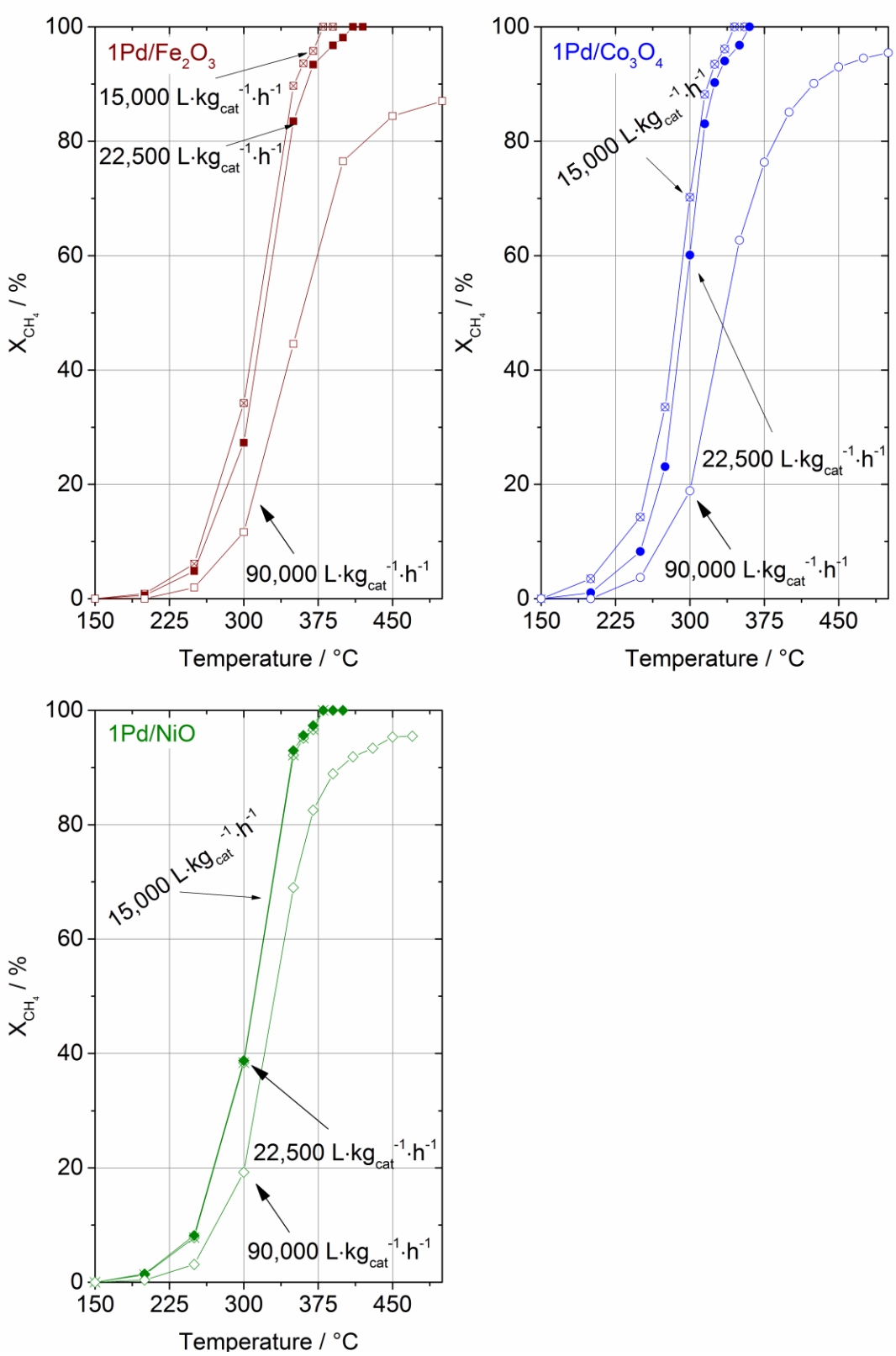

Figure S1. Catalytic activity tests for methane combustion, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, catalyst masses: 300, 200 and 50 mg, catalysts: 1Pd/Fe_2O_3, 1Pd/Co_3O_4 and 1Pd/NiO.

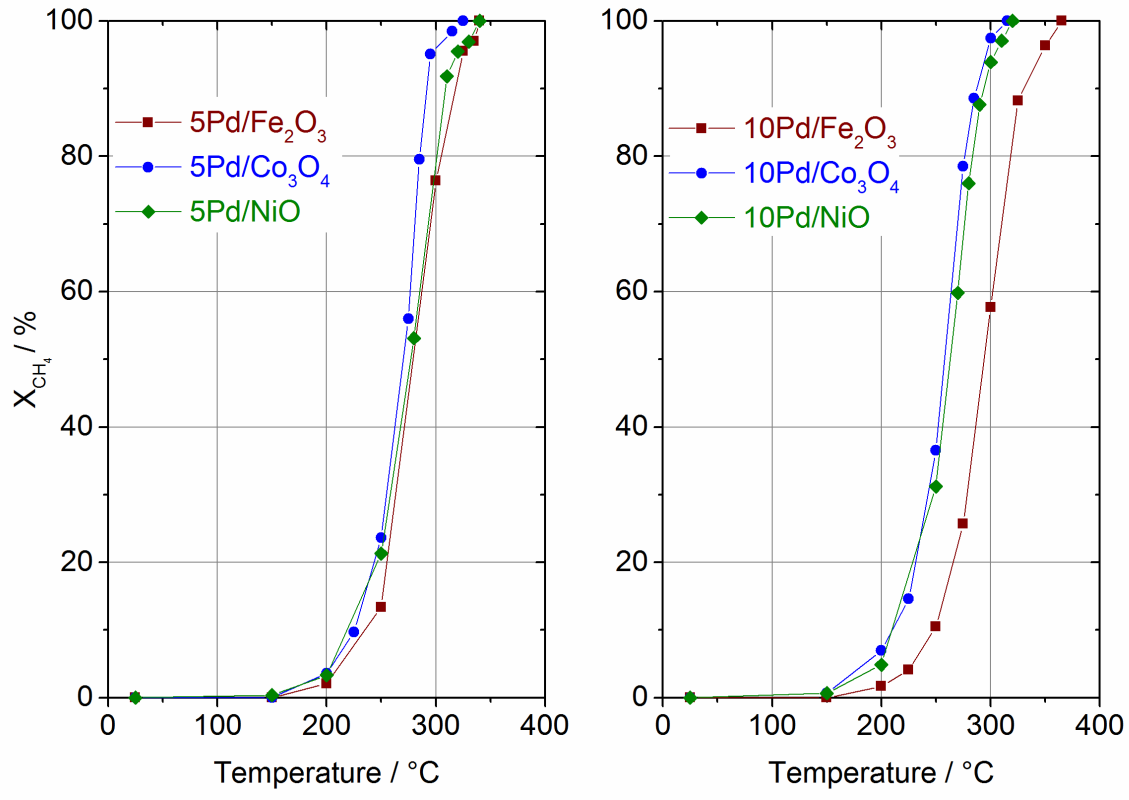

Figure S2. Catalytic activity tests for methane combustion, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, at 22.500 L·kg^{-1}·h^{-1}: (left) 5Pd/Fe_2O_3, 5Pd/Co_3O_4, 5Pd/NiO (right) 10Pd/Fe_2O_3, 10Pd/Co_3O_4, 10Pd/NiO.

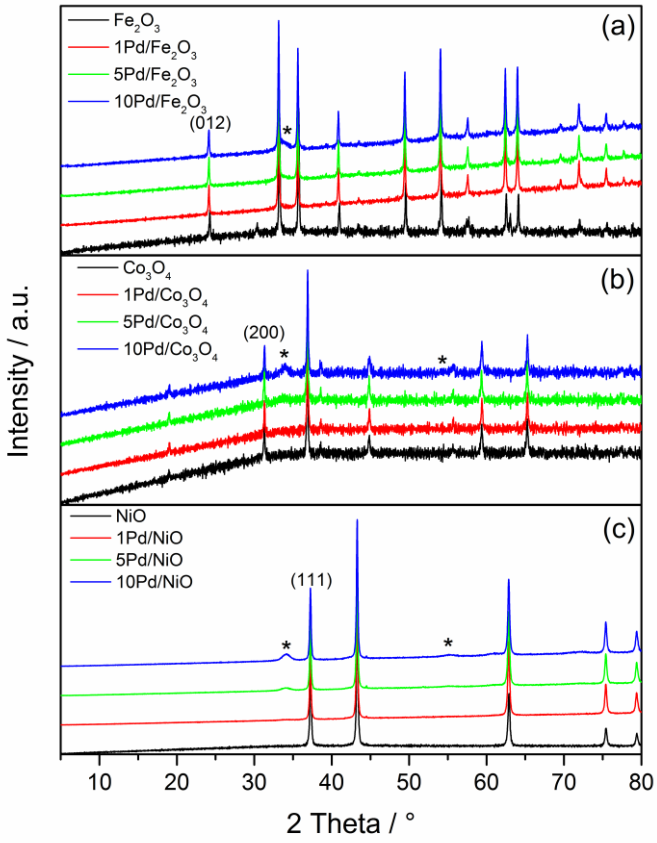

Figure S3. XRD Diffraction patterns of (a) Fe_2O_3, 1Pd/Fe_2O_3, 5Pd/Fe_2O_3, 10Pd/Fe_2O_3, (b) Co_3O_4, 1Pd/Co_3O_4, 5Pd/Co_3O_4, 10Pd/Co_3O_4 and (c) NiO, 1Pd/NiO, 5Pd/NiO, 10Pd/NiO; * PdO reference (ICSD 24692, [1]).

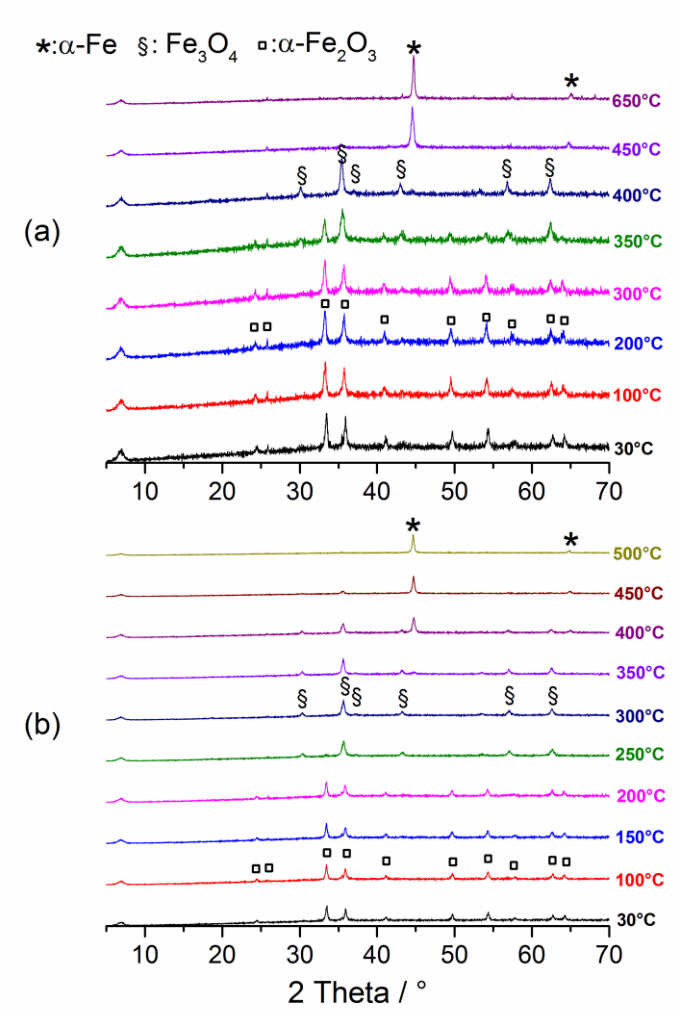

Figure S4. *In situ* XRD under H_2 reduction (a) Fe_2O_3 and (b) 1Pd/Fe_2O_3, 5 vol% H_2 in He, 10 ml·min^{-1} with a heating rate of 10 K/min (slightly higher phase transition temperatures are specific for the method).

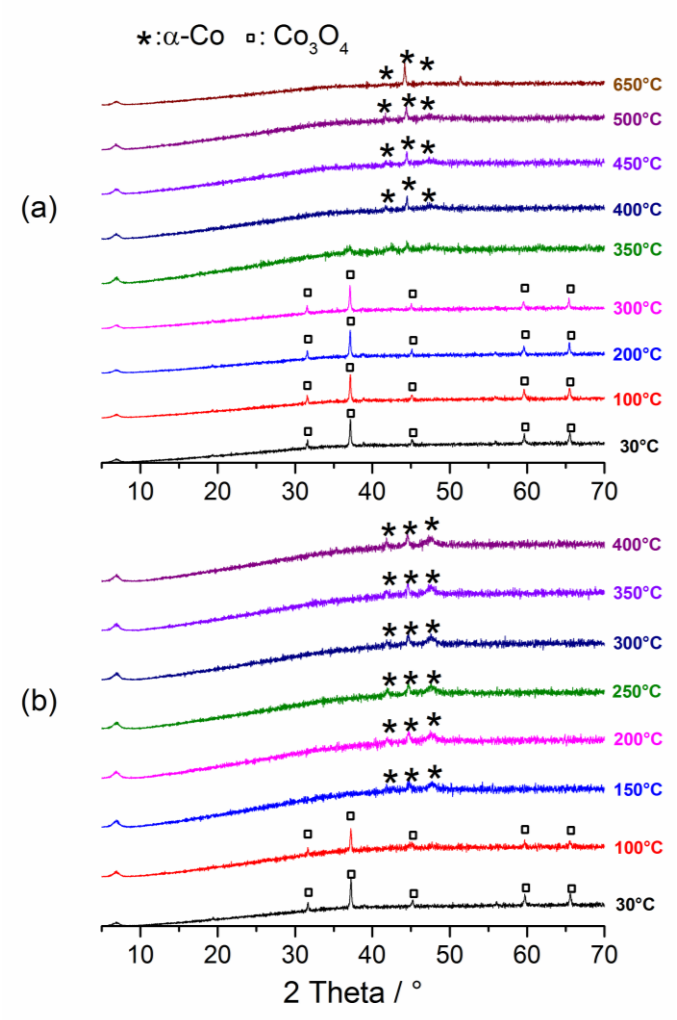

Figure S5. *In situ* XRD under H_2 reduction (a) Co_3O_4 and (b)1Pd/Co_3O_4, 5 vol% H_2 in He, 10 ml·min^{-1} with a heating rate of 10 K/min (slightly higher phase transition temperatures are specific for the method).

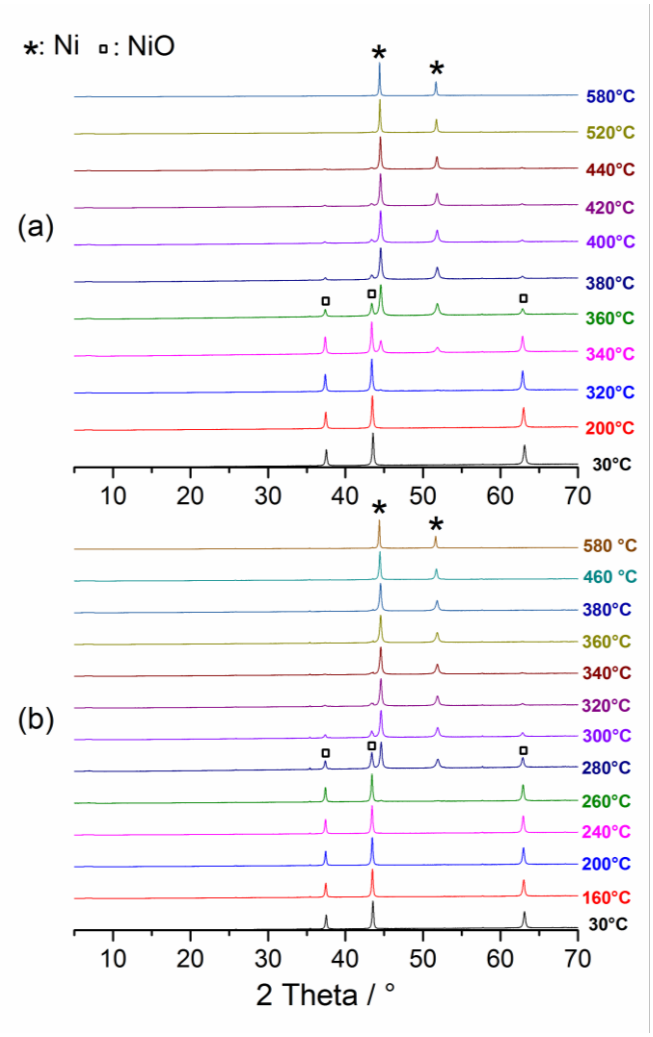

Figure S6. *In situ* XRD under H_2 reduction (a) NiO and (b)1Pd/NiO, 5 vol% H_2 in He, 10 ml/min with a heating rate of 10 K/min (slightly higher phase transition temperatures are specific for the method).

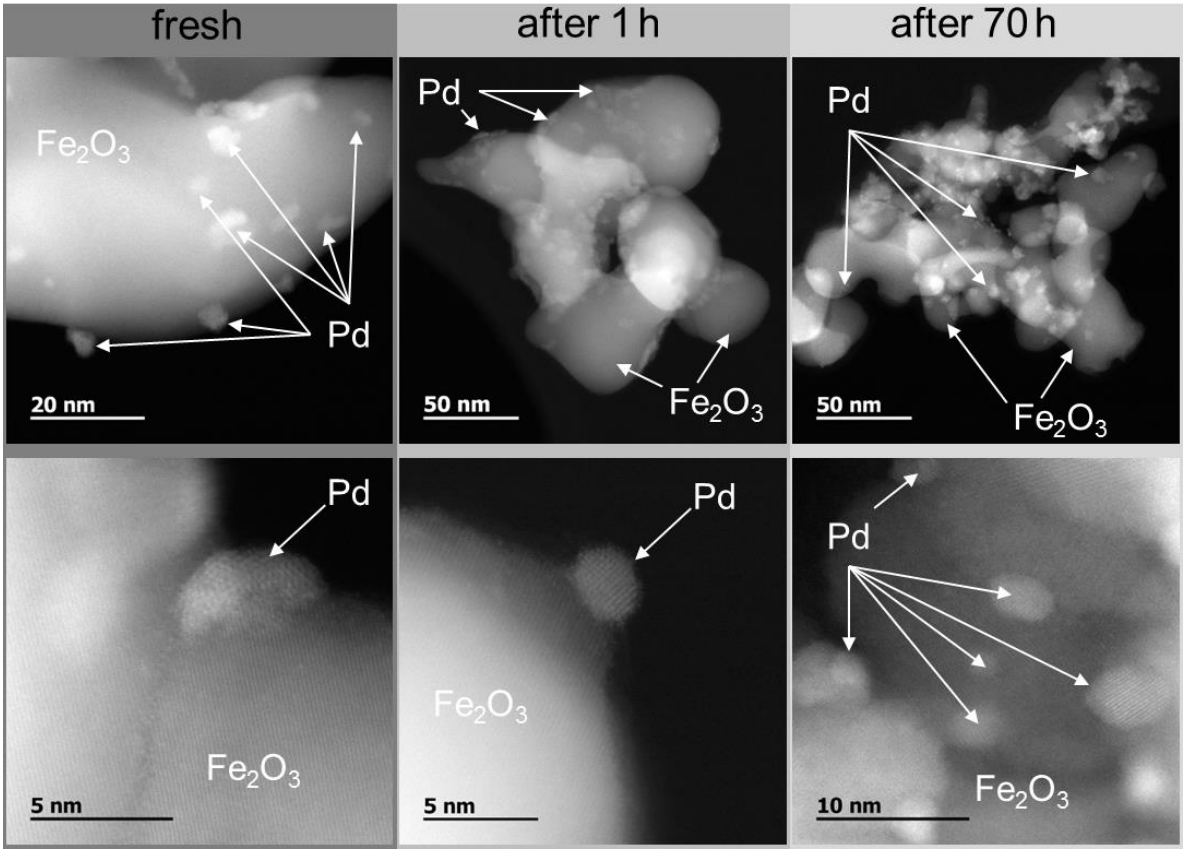

Figure S7. HAADF-STEM images of fresh and spent samples after long-term tests (at T_{100}, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, at 22,500 $L \cdot kg^{-1} \cdot h^{-1}$): 1Pd/$Fe_2O_3$.

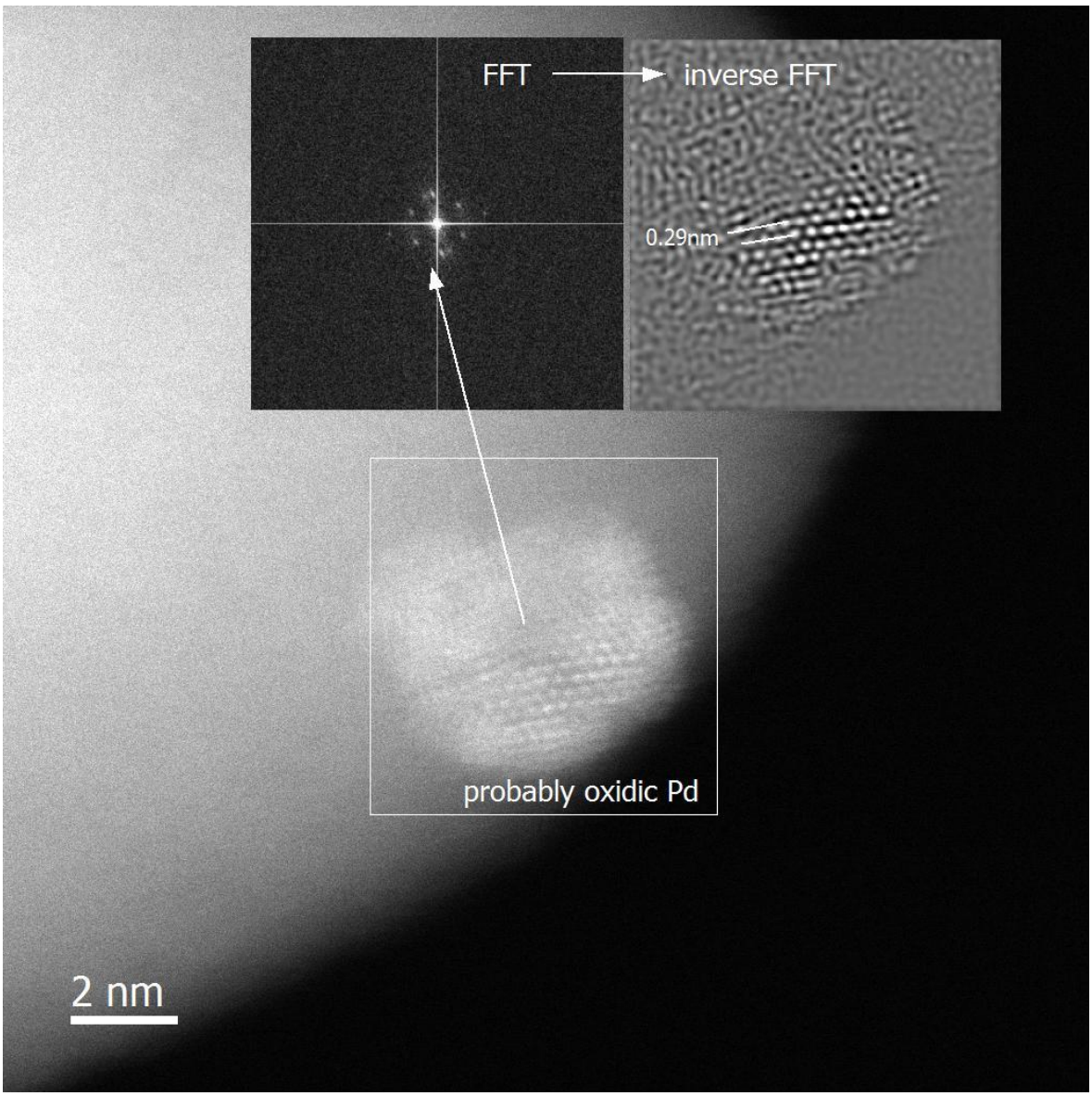

Figure S8. HAADF-STEM, FFT and inverse FFT of fresh 1Pd/Fe_2O_3.

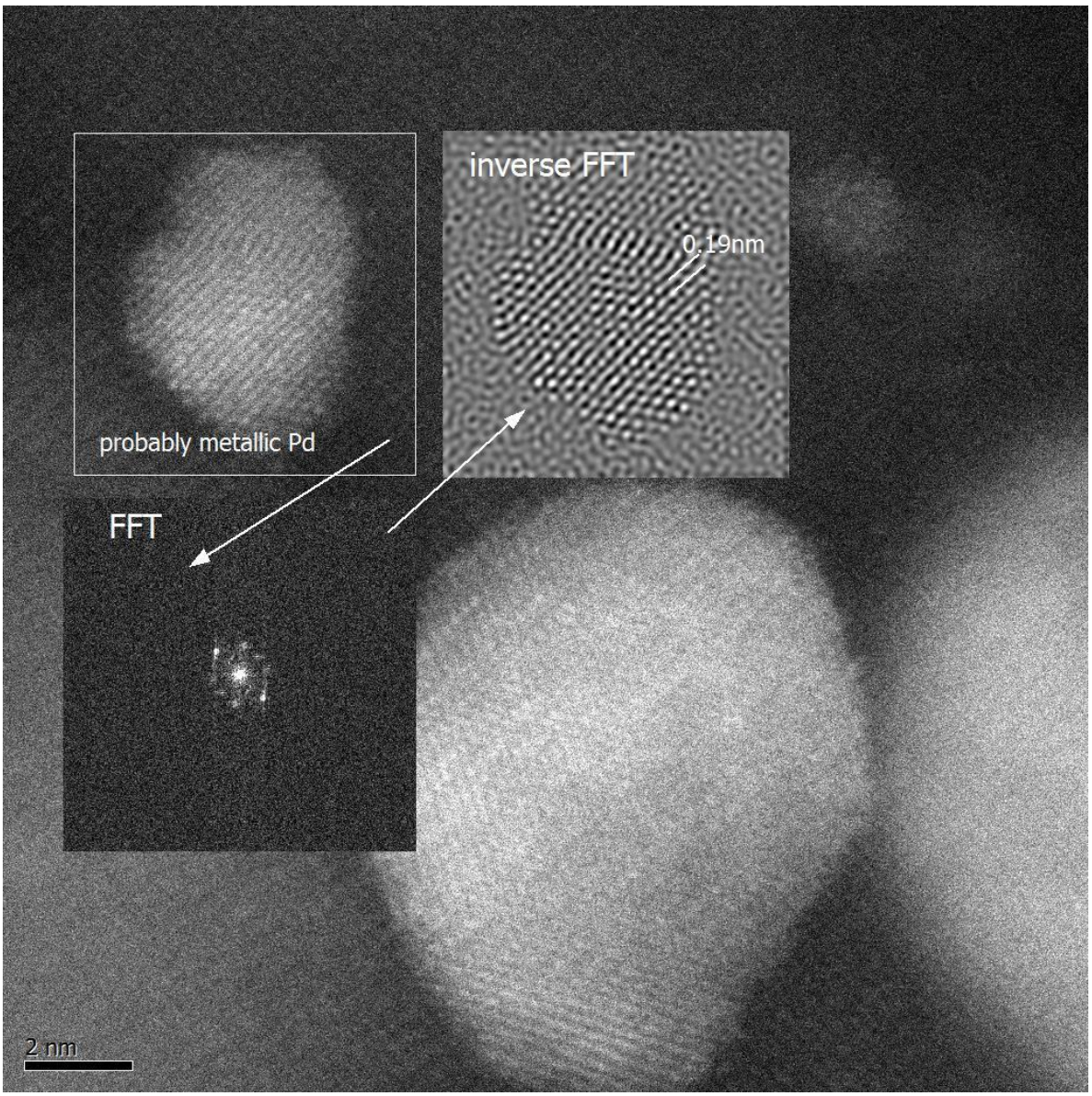

Figure S9. HAADF-STEM, FFT and inverse FFT of 1Pd/Fe_2O_3 after 1h on stream.

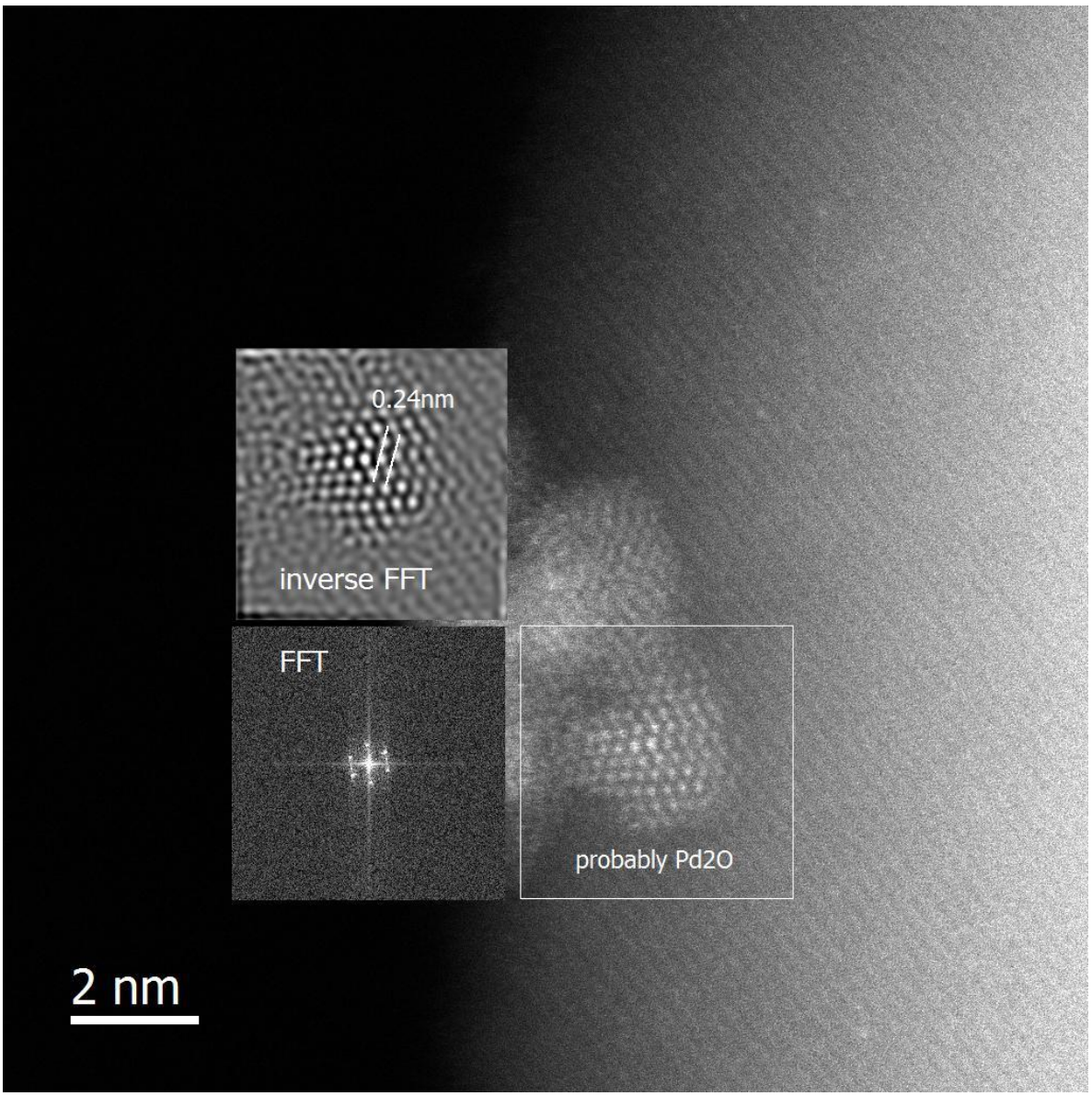

Figure S10. HAADF-STEM, FFT and inverse FFT of 1Pd/Fe_2O_3 after 1h on stream.

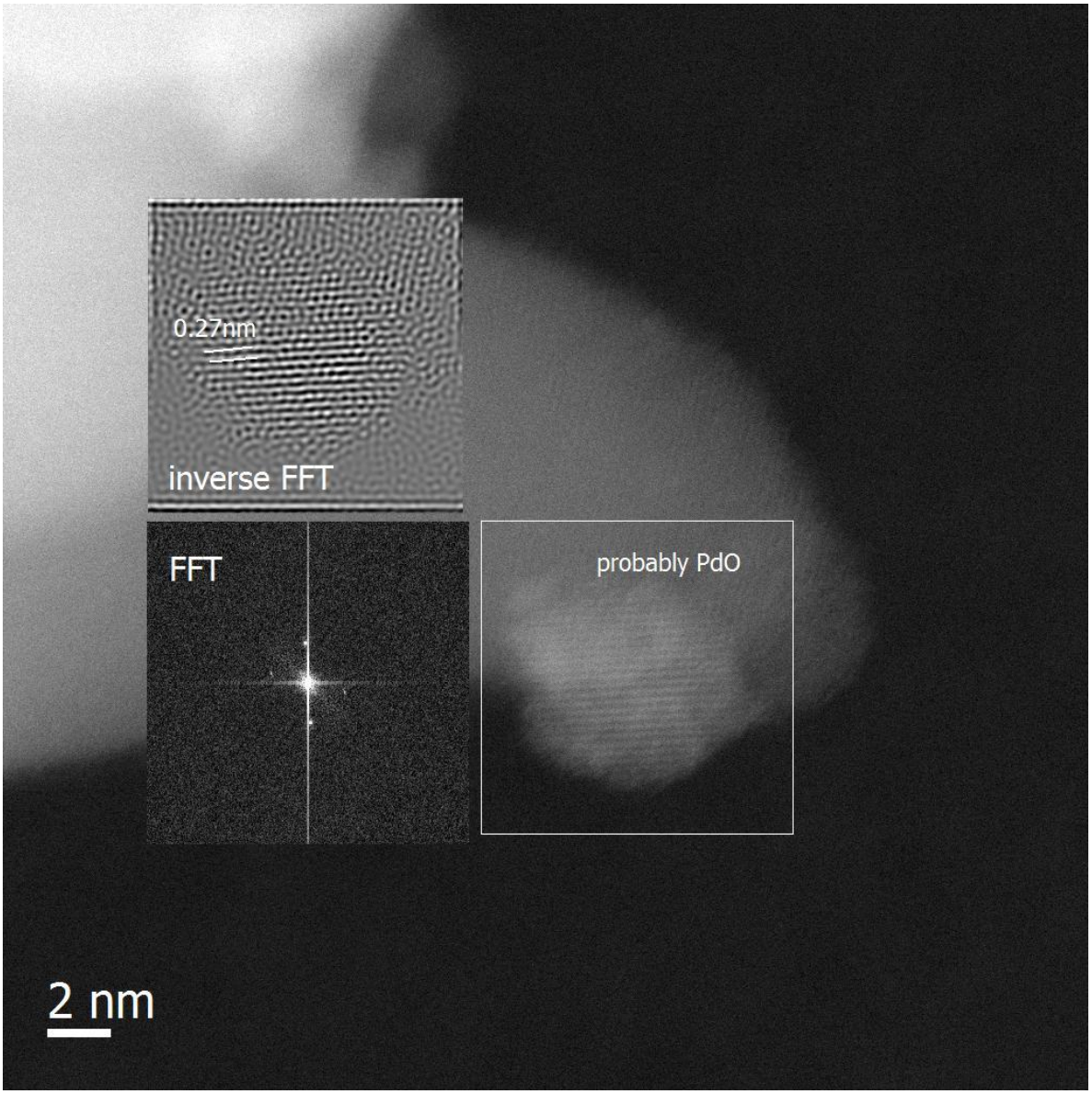

Figure S11. HAADF-STEM, FFT and inverse FFT of 1Pd/Fe_2O_3 after 70 h on stream.

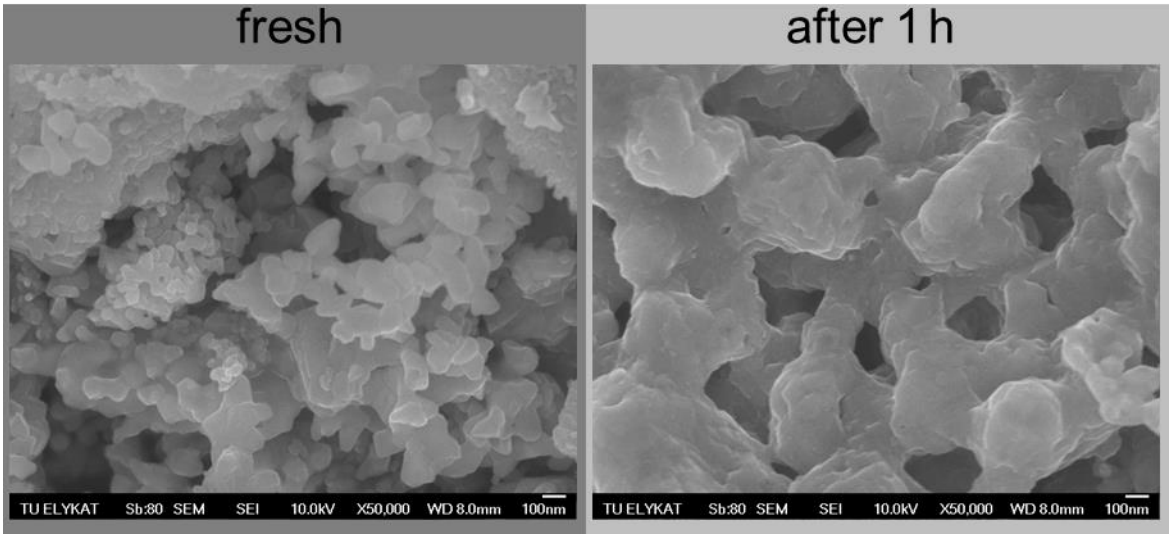

Figure S12. SEM images of fresh and spent samples after 1 h of the long-term tests (at T_{100}, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, at 22,500 $L \cdot kg^{-1} \cdot h^{-1}$): 1Pd/$Co_3O_4$.

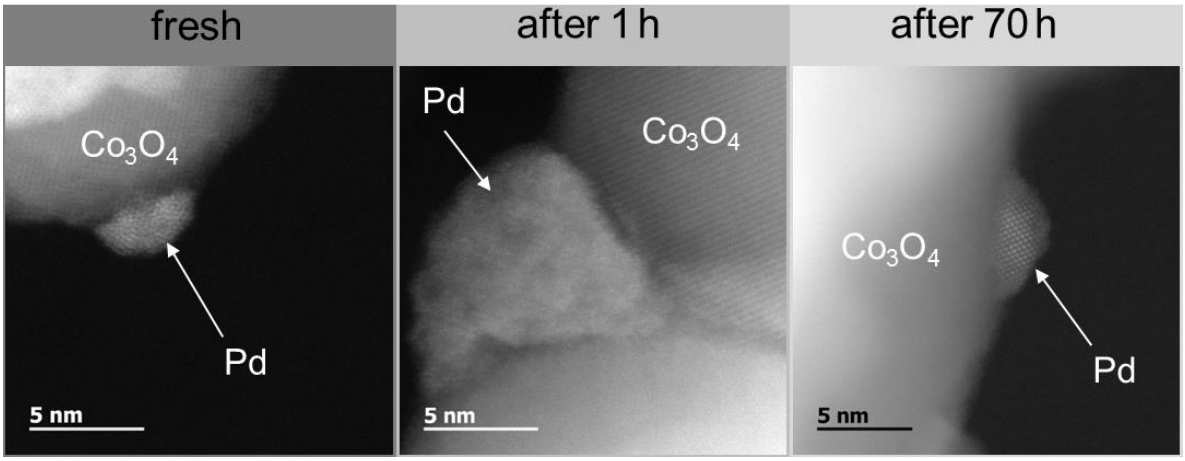

Figure S13. HAADF-STEM images of fresh and spent samples after long-term tests (at T_{100}, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, at 22,500 $L \cdot kg^{-1} \cdot h^{-1}$): 1Pd/$Co_3O_4$.

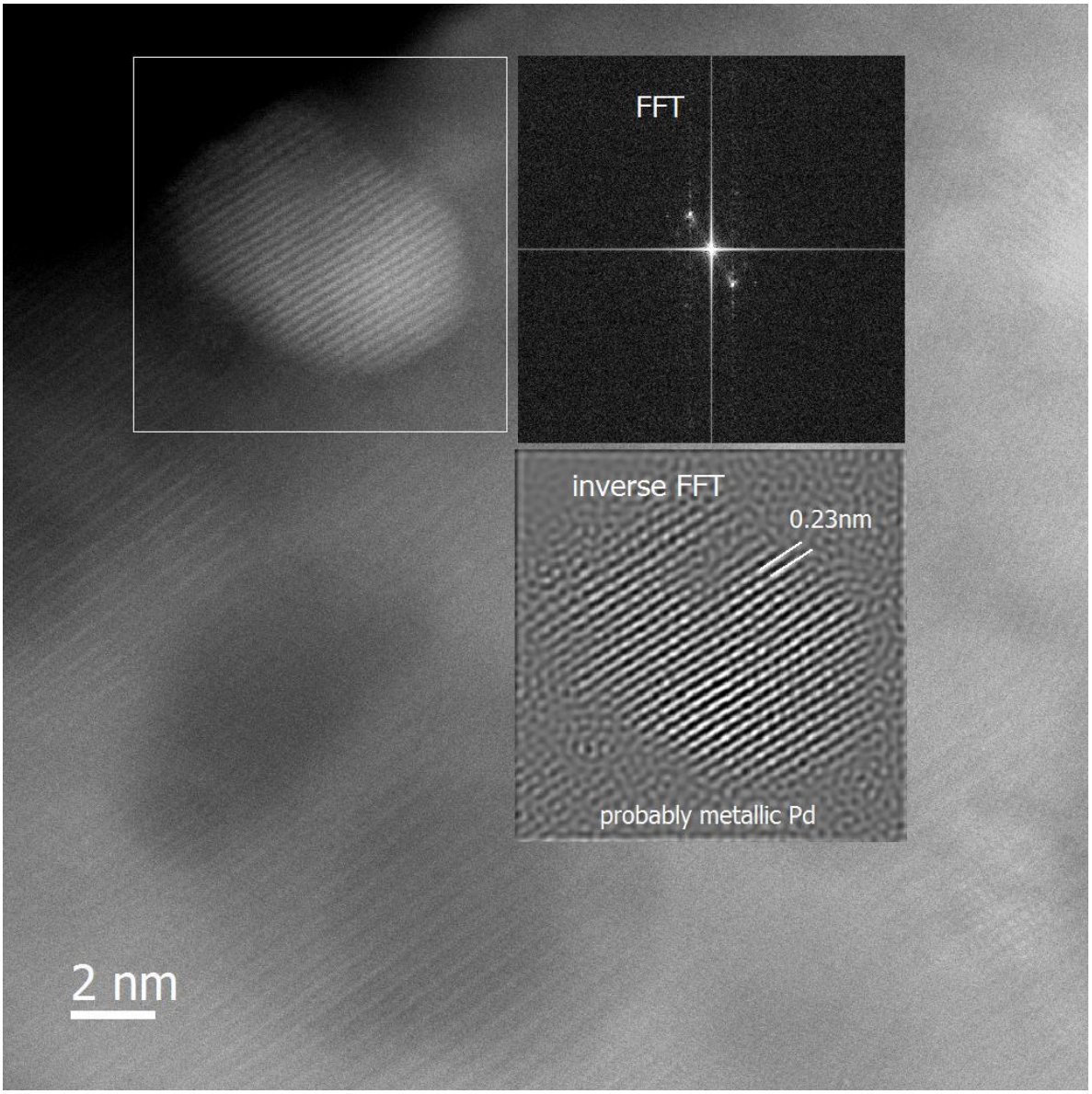

Figure S14. HAADF-STEM, FFT and inverse FFT of 1Pd/Co_3O_4 after 1 h on stream.

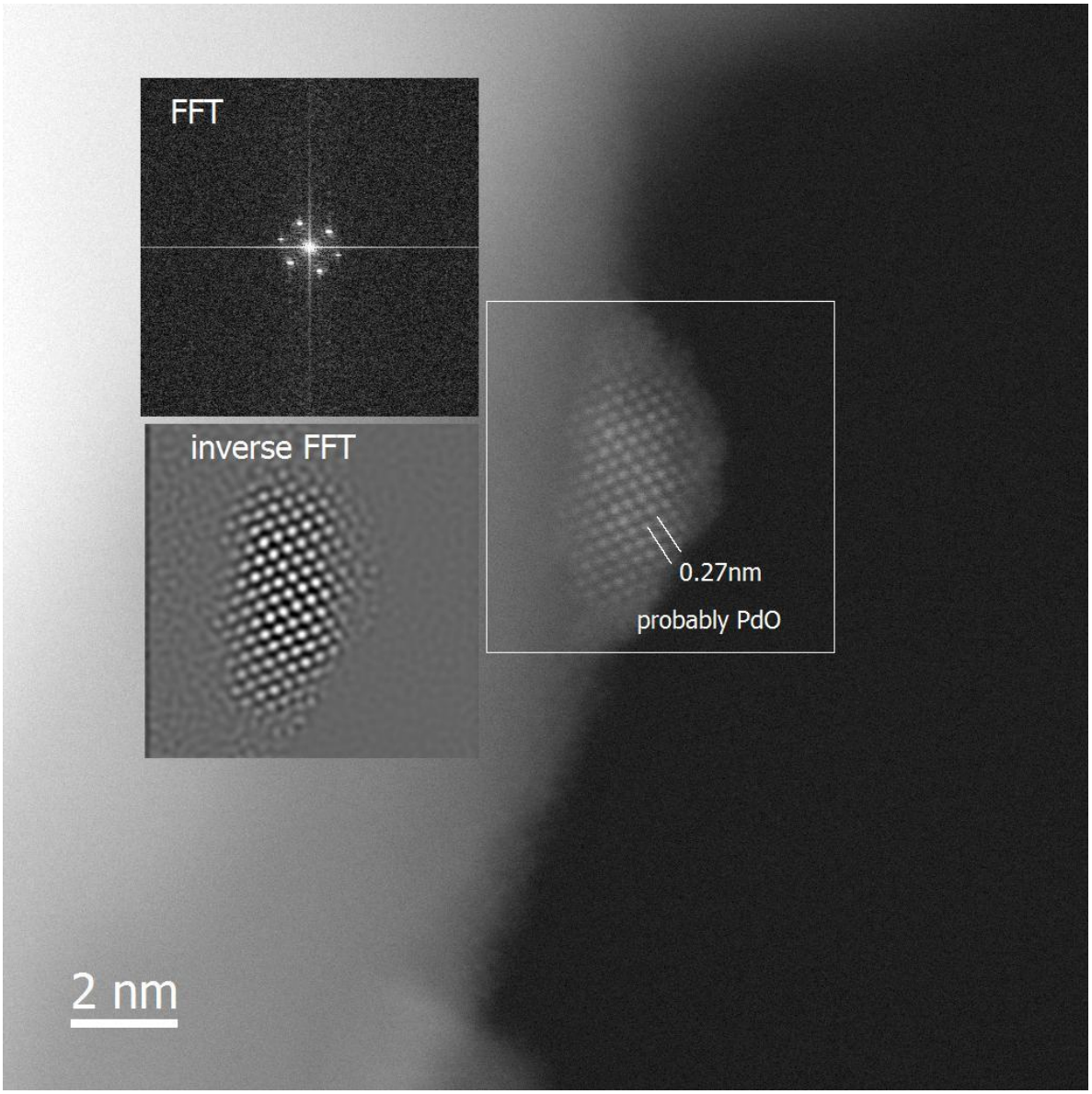

Figure S15. HAADF-STEM, FFT and inverse FFT of 1Pd/Co_3O_4 after 70 h on stream.

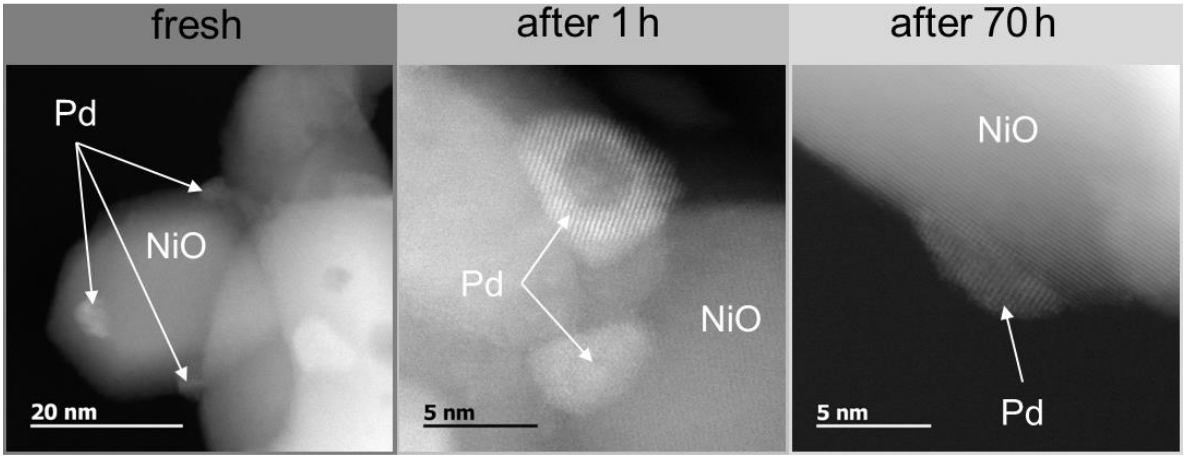

Figure S16. HAADF-STEM images of fresh and spent samples after long-term tests (at T_{100}, feed gas consisting of CH_4:O_2:N_2 = 1:18:81, at 22,500 $L \cdot kg^{-1} \cdot h^{-1}$): 1Pd/NiO.

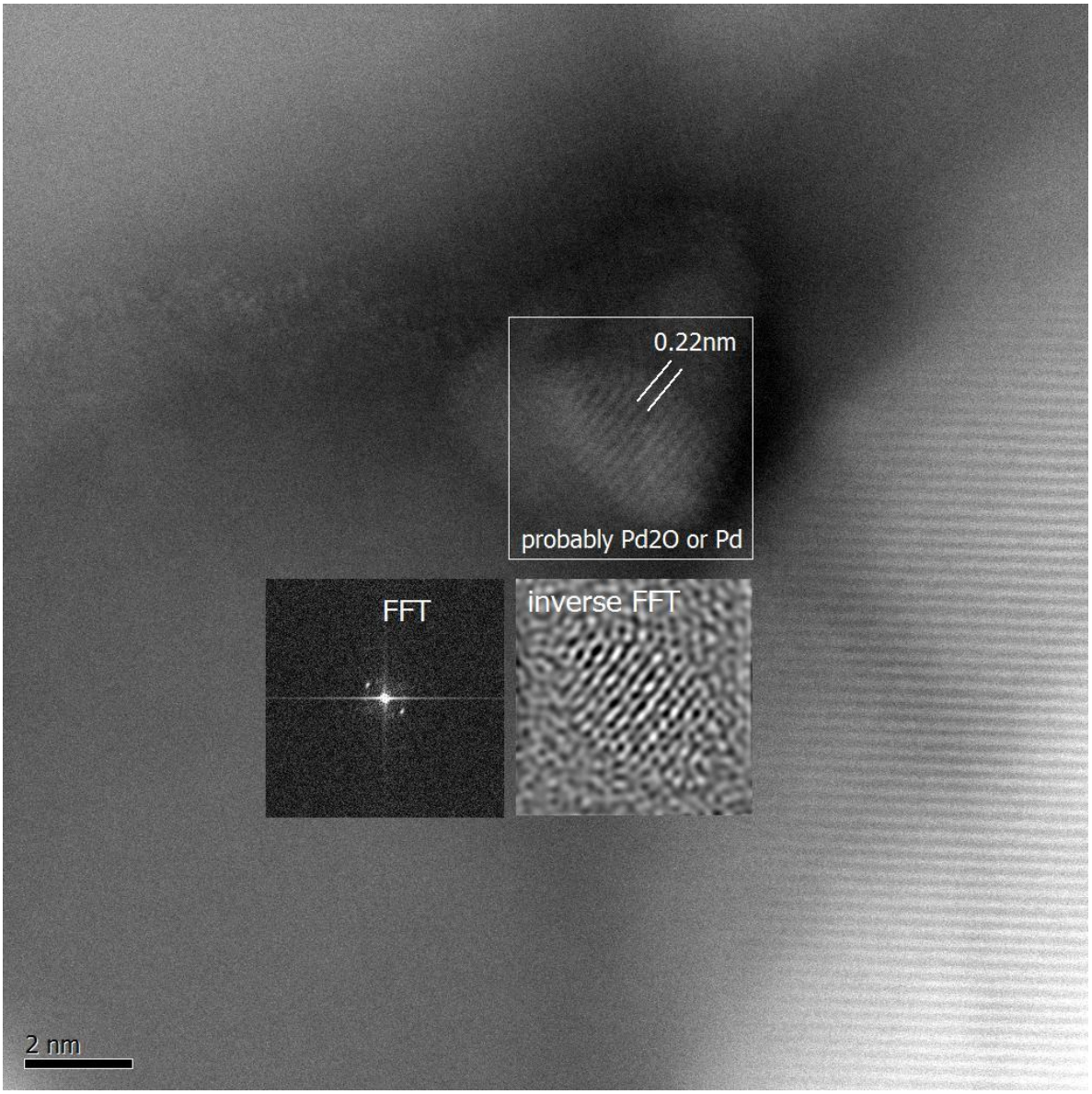

Figure S17. HAADF-STEM, FFT and inverse FFT of 1Pd/NiO after 1 h on stream.

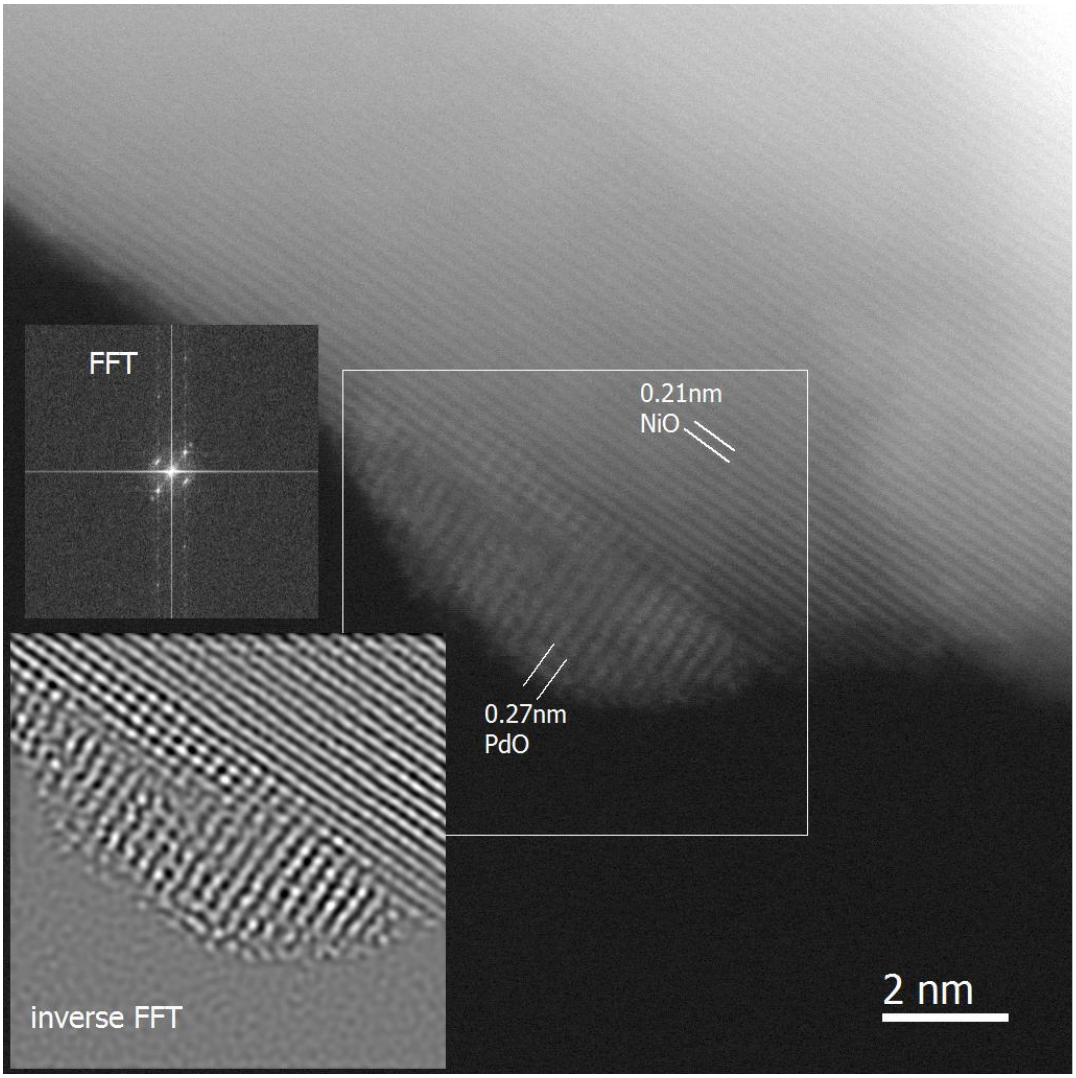

Figure S18. HAADF-STEM, FFT and inverse FFT of 1Pd/NiO after 70 h on stream.

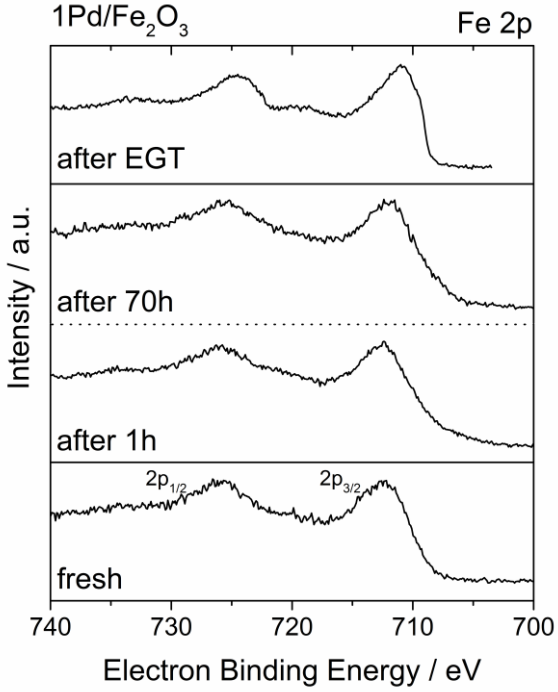

Figure S19. 1Pd/Fe_2O_3 - XPS spectra of fresh and spent samples, after 1 h, 70 h and after test with exhaust gas (EGT).

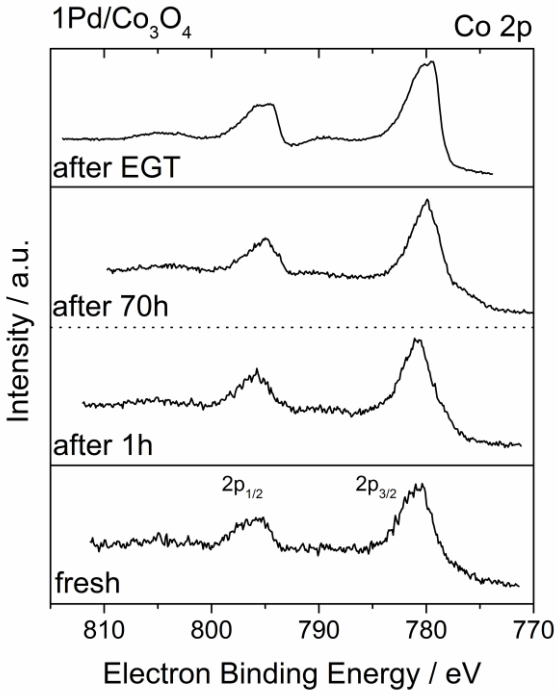

Figure S20. 1Pd/Co_3O_4 - XPS spectra of fresh and spent samples, after 1 h, 70 h and after test with exhaust gas (EGT).

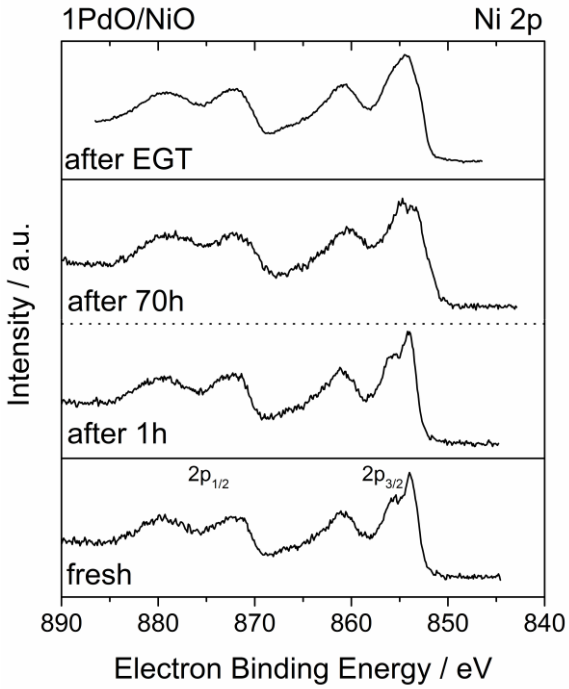

Figure S21. 1Pd/NiO - XPS spectra of fresh and spent samples, after 1 h, 70 h and after test with exhaust gas (EGT).

References

Waser, J., H.A. Levy, and S.W. Peterson, *The structure of PdO.* Acta Crystallographica, 1953. **6** (7): p. 661-663.