

Kinetics of low-temperature methane oxidation over SiO₂ encapsulated bimetallic Pd-Pt nanoparticles – Supplementary Information

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1. Calculation of silica shell porosity

The porosity of the shell can be estimated from the data using the parallel pore model, which is valid for a narrow pore size distribution. We note that:

$$d_P = \frac{4\varepsilon}{S_A \rho_S (1-\varepsilon)}$$

From Table 1 of the main text, the mean pore diameter of the shell was 3.4 nm, the surface area was 592 m²/g, and we take the density of the non-porous shell material as 2200 kg/m³.

Substitute:

$$3.4 \times 10^{-9} \text{ m} = \frac{4\varepsilon}{592,000 \frac{\text{m}^2}{\text{kg}} 2200 \frac{\text{kg}}{\text{m}^3} (1-\varepsilon)}$$

Solving for the porosity gives a value of 52.5%.

2. Calculation of the average number of cores in a shell

On average, there are multiple cores per shell. The average number of cores can also be calculated (estimated) from the physical data. The mass percent of PGM in the catalyst calculated from the NAA analysis was 11.19%. The number of cores then depends on the relative densities of the cores and shell. The mass fraction of PGM is given by the formula:

$$X_{PGM} = 0.1119 = \frac{n D_C^3 \rho_C}{n D_C^3 \rho_C + (D_S^3 - D_C^3) \rho_S (1-\varepsilon)}$$

Substitute the numbers:

$$0.1119 = \frac{n 9^3 16,714}{n 3.4^3 16,714 + (70^3 - n 9^3) 2200 (1-0.525)}$$

Solve for $n=3.7$. On average there should be about 3.7 cores per shell.

3. Predicted and experimental ignition curves, Model 1 for dry combustion

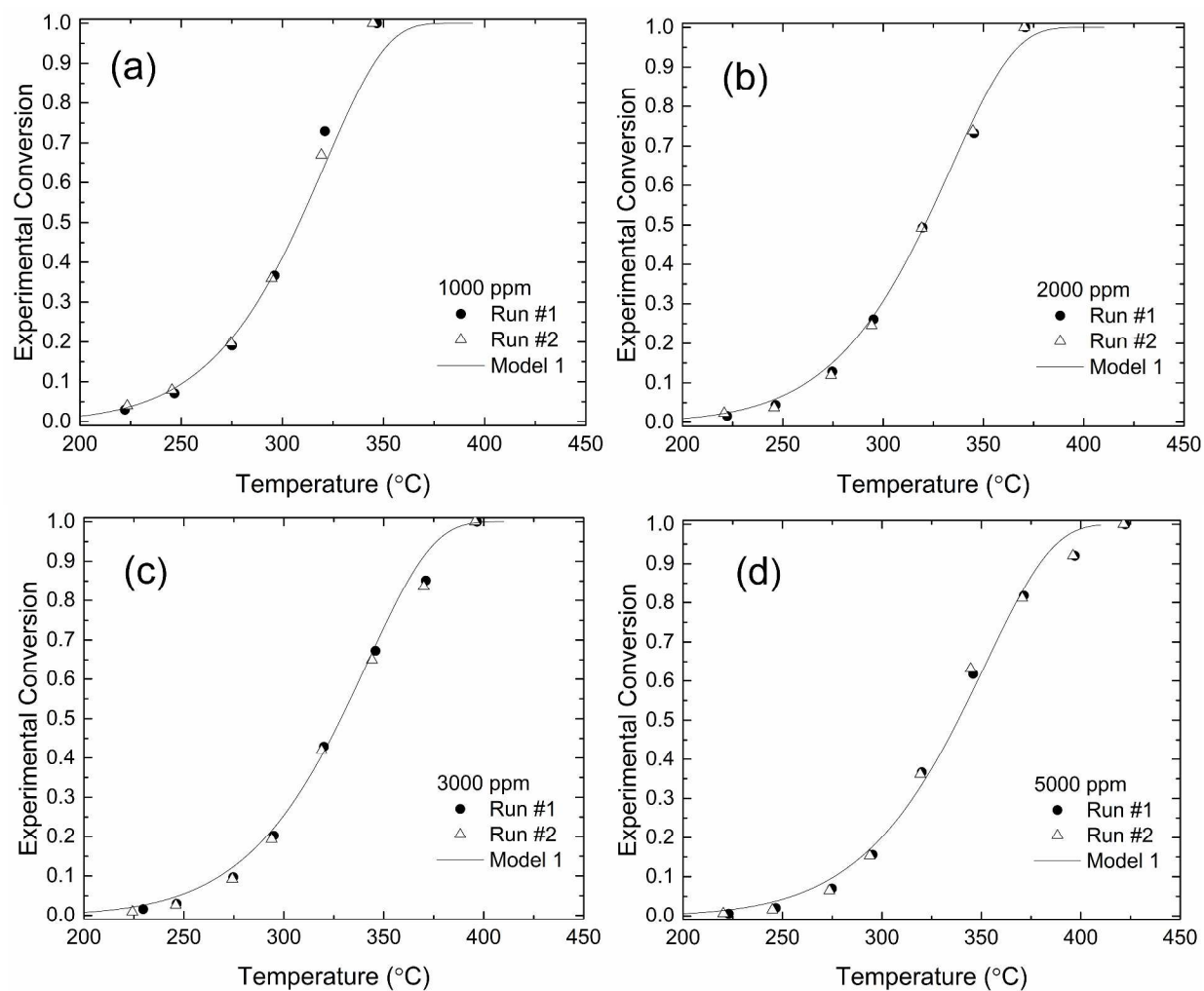


Figure S1. The predicted ignition curve produced by Model 1 and the experimental points. Reaction in dry conditions at initial CH_4 concentrations of a) 1000 ppm, b) 2000 ppm, c) 3000 ppm and d) 5000 ppm.

4. Predicted and experimental ignition curves, Model 2 for wet combustion

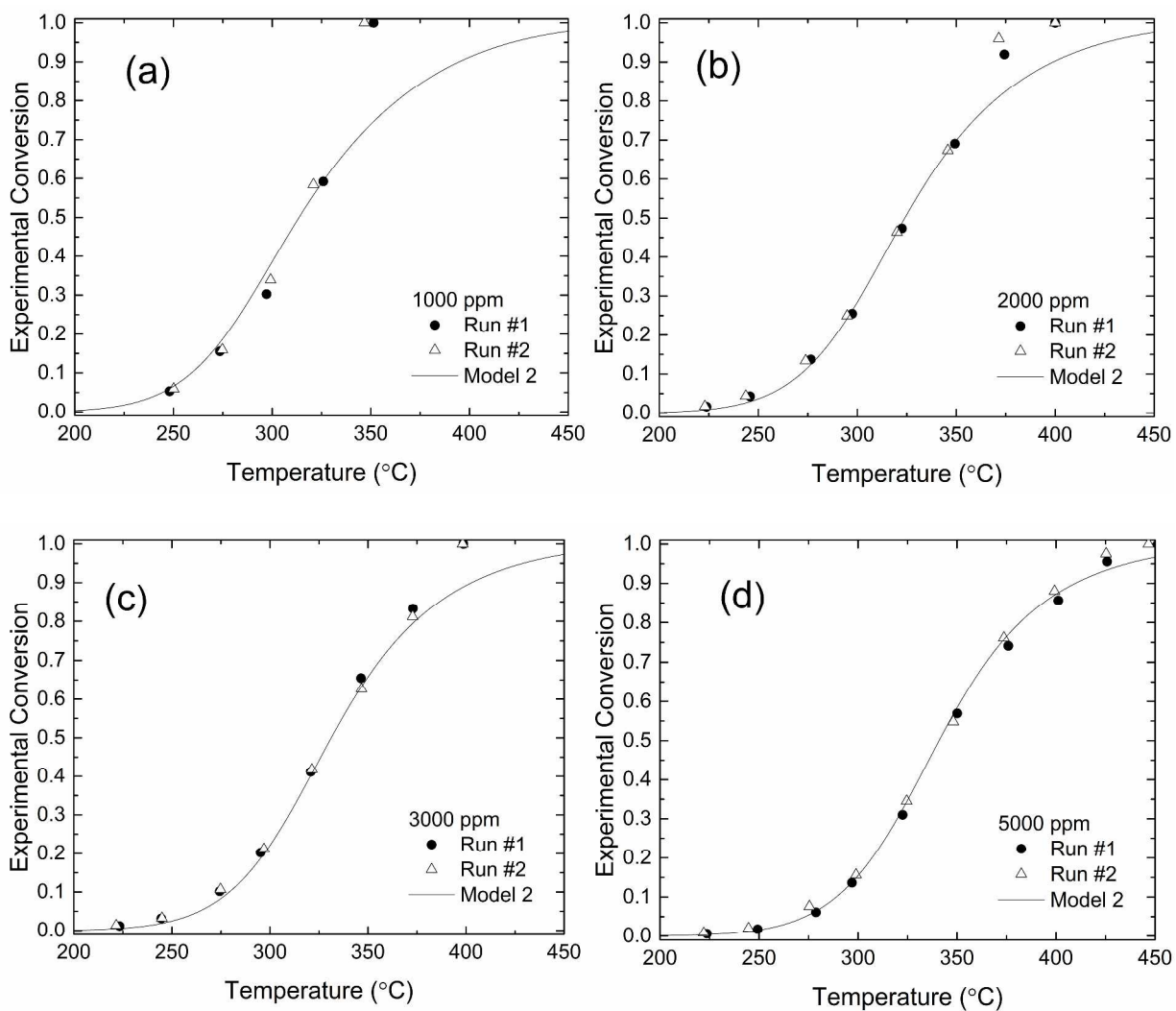


Figure S2. The predicted light-off curve produced by Model 2 and the experimental points. Reaction in the presence of 5% added H₂O and initial CH₄ concentrations of a) 1000 ppm, b) 2000 ppm, c) 3000 ppm and d) 5000 ppm.