

The solvothermal formation mechanism of Pd-containing bimetals

A. B. Borup^{1*}, M. Kløve¹, A. D. Bertelsen¹, B. B. Iversen¹

¹Center for Integrated Materials Research, Department of Chemistry, Aarhus University, Langelandsgade 140, 8000 Aarhus,, Denmark

*Contact: andersborup@chem.au.dk

Pd is commonly used as a catalyst in a variety of fields including alcohol oxidation [1], selective hydrogenation/dehydrogenation [2, 3], and direct synthesis of H₂O₂ [4]. To improve the catalytic performance and abilities of the Pd catalyst, alloying Pd with another element is commonly used since alloying can give rise to three effects; the element can change the electronic nature of the catalytic active atom, it can block neighboring catalytic sites, which alters the bond to poisonous intermediates, and lastly, the metal can catalyze the reaction by itself [5].

However, there exists no general theory for how bimetallic nanoparticles are formed under solvothermal conditions, although they are commonly believed to form as a result of a co-reduction, which was shown for the Pb_xPd_y case [8]. In addition to this, the solvent can also be used to control the amount of the secondary element incorporated into the Pd-structure, with, at least for the Sn_xPd_y case, ethanol giving a significantly more Pd-poor phase at the end of the *in situ* experiment compared with ethylene glycol.

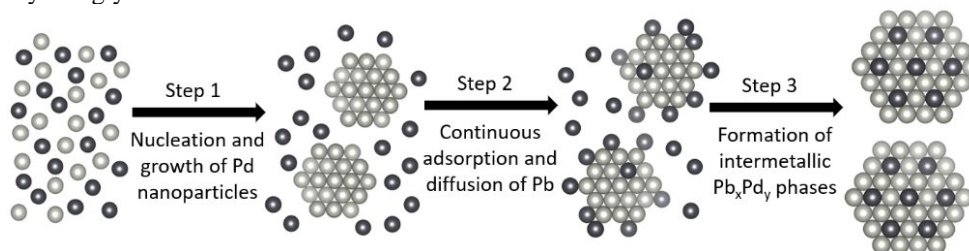


Figure 1. Illustration of the proposed mechanism for the formation of intermetallic Pb_xPd_y nanoparticles. The light grey atoms are Pd, while the dark grey atoms are Pb. For simplicity, ions and atoms are shown by the same symbols, and counter ions are omitted. Reproduced from Borup *et al.* [8]

- [1] P. Xin, J. Li, Y. Xiong, X. Wu, J. Dong, W. Chen, Y. Wang, L. Gu, J. Luo, H. Rong, C. Chen, Q. Peng, D. Wang & Y. Li. (2018). *Angew. Chem. Int. Ed.* **57**, 4642-4646.
- [2] Y. Izawa, D. Pun, & S. S. Stahl. (2011). *Science*. **333**, 209-213.
- [3] S. Kidambi, J. Dai, J. Li & M. L. Bruening. (2004). *J. Am. Chem. Soc.* **126**, 2658-2659.
- [4] Q. Liu, J. C. Bauer, R. E. Schaak, & J. H. Lundsford. (2008). *Angew. Chem. Int. Ed.* **47**, 6221-6224.
- [5] S. Mondal, V. S. K. Choutipalli, B. K. Jena, V. Subramanian & C. R. Raj. (2020). *J. Phys. Chem C.* **124**, 9631-9643.
- [6] D. Saha, E. D. Bøjesen, K. M. Ø. Jensen, A.-C. Dippel & B. B. Iversen. (2015). *J. Phys. Chem. C.* **119**, 13357-13362.
- [7] D. Saha, E. D. Bøjesen, A. H. Mamakhel, M. Bremholm & B. B. Iversen (2017). *ChemNanoMat.* **3**, 472-478.
- [8] A. B. Borup, A. D. Bertelsen, M. Kløve, R. S. Christensen, N. L. N. Broge, A.-C. Dippel, M. R. V. Jørgensen & B. B. Iversen. (2023). *Nanoscale.* **15**, 18481-18488.
- [9] N. L. N. Broge, A. D. Bertelsen, I. G. Nielsen, M. Kløve, M. Roelsgaard, A.-C. Dippel, M. R. V. Jørgensen & B. B. Iversen. (2024). *Phys. Chem. Chem. Phys.* **26**, 12121-12132.
- [10] N. L. N. Broge, F. Søndergaard-Pedersen, M. Roelsgaard, X. Hassing-Hansen & B. B. Iversen. *Nanoscale.* **12**, 8511-8518.