

Supporting Information for

Fractal growth of platinum electrodeposits revealed by *in situ* electron microscopy

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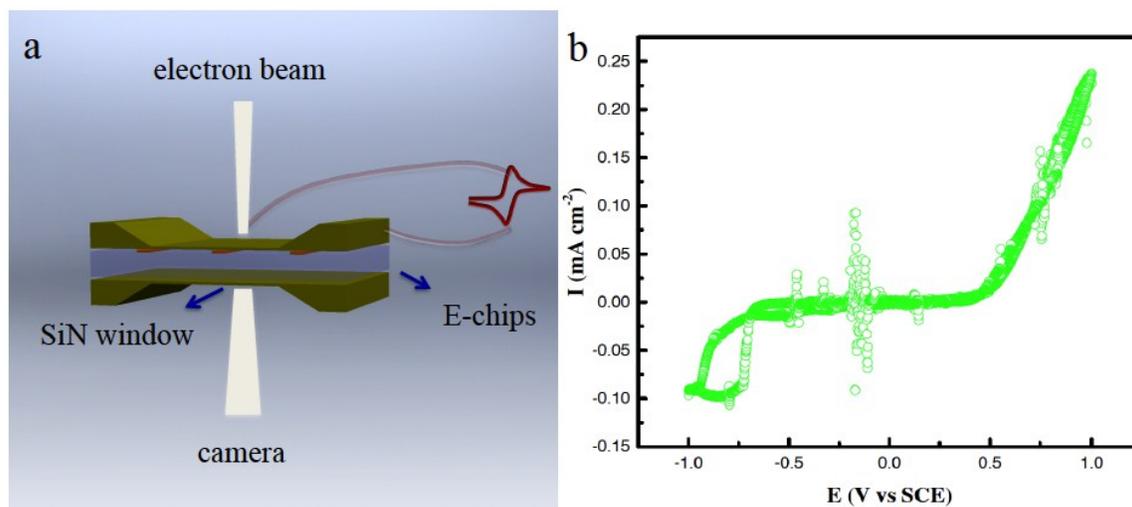


Figure S1. Experimental setup. (a), Cross-sectional schematic of the liquid cell. Electrolyte is encapsulated between two SiN membranes. Three electrodes on the SiN window are connected to the electrical circuit. (b), CV profile for the as-assembled liquid cell.

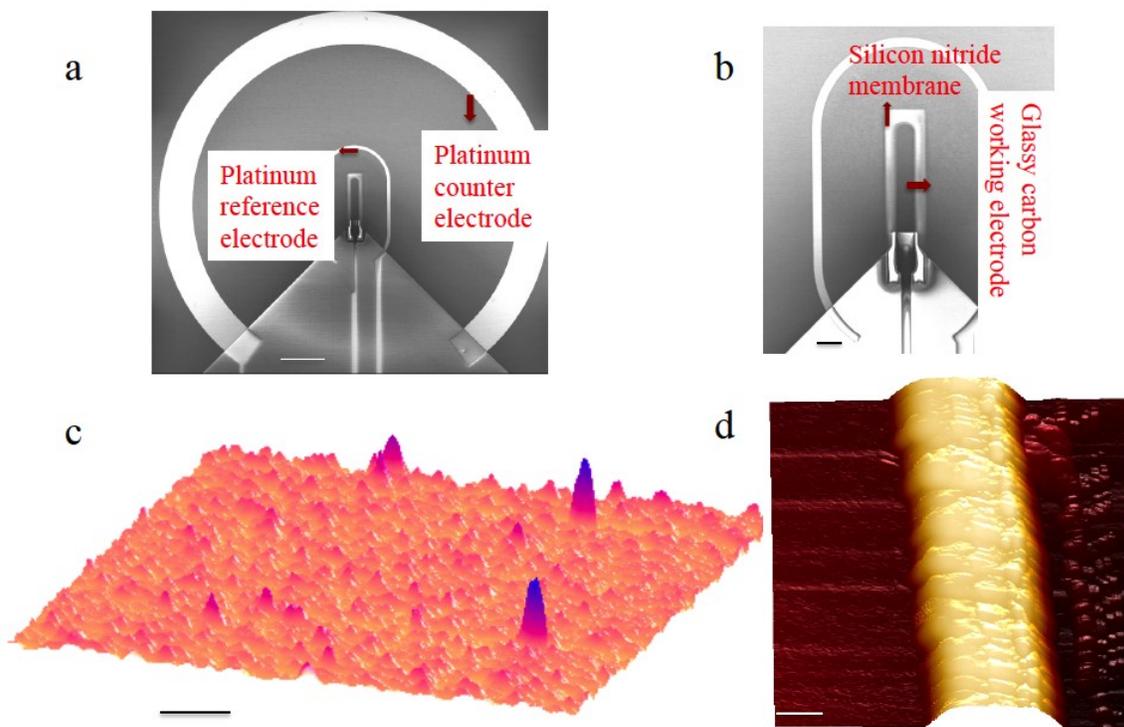


Figure S2. Characterization of electrochemical cell. (a), (b) SEM images of E-chip with three electrodes on top. Glassy carbon sitting in the middle serves as the working electrode. Scale bar: 200 μm in a, 200 nm in b. (c), (d) Atomic force microscopy (AFM) characterization of the working electrode. Scale bar in c and d are 50 nm and 20 nm, respectively.



Figure S3. Schematic of equipotential lines around a bump.

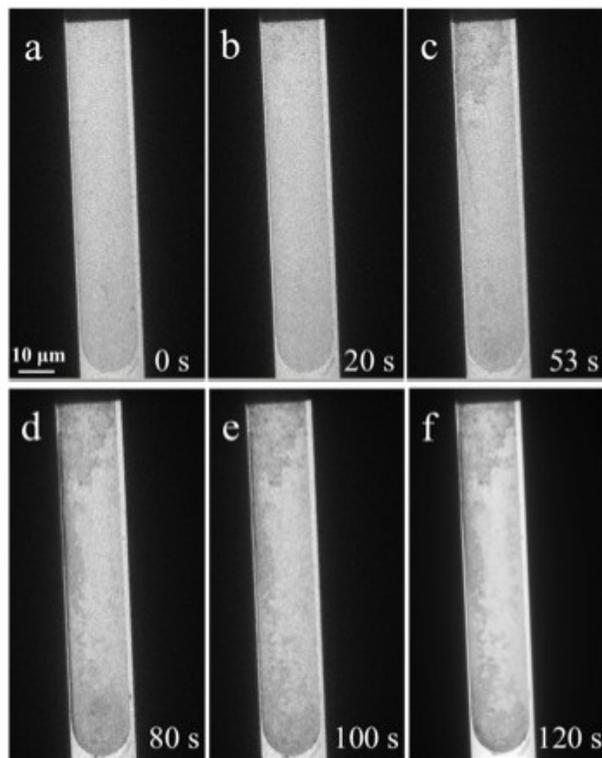


Figure S4. Overview of the electrochemical deposition process. (a), Transparent SiN_x window is sitting in the middle of the chip; the circular edge shows glassy carbon electrode which covers most of the window. (b-f), After potentiostatic signal applied to the chip, dark deposits show up and gradually cover the glassy carbon, especially edge areas. The electrolyte concentration is 0.1 mol/L.

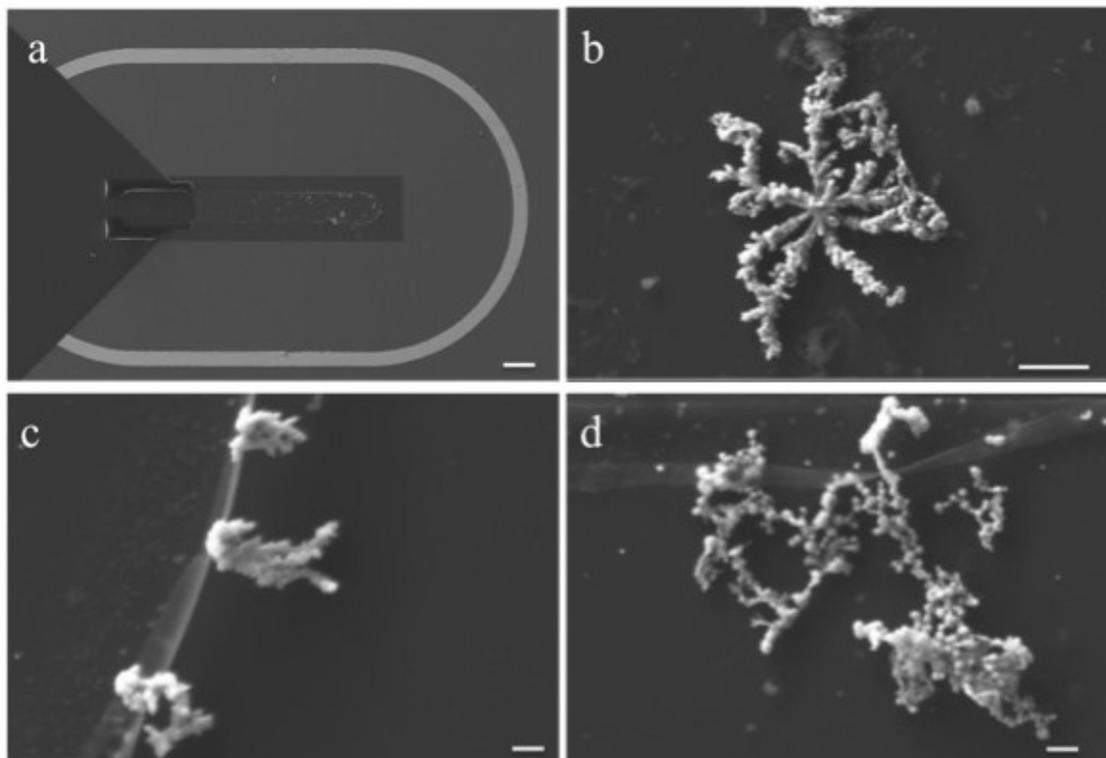


Figure S5. *ex situ* SEM images of Pt deposits. (a), SEM image of the disassembled E-chip with the glassy carbon as deposition electrode in the middle and platinum as counter electrode in the out circle after electrodeposition. Deposits cover most areas of the glassy carbon. (b-d), SEM images of the deposit in different areas showing typical fractal featured flakes and filament morphologies.

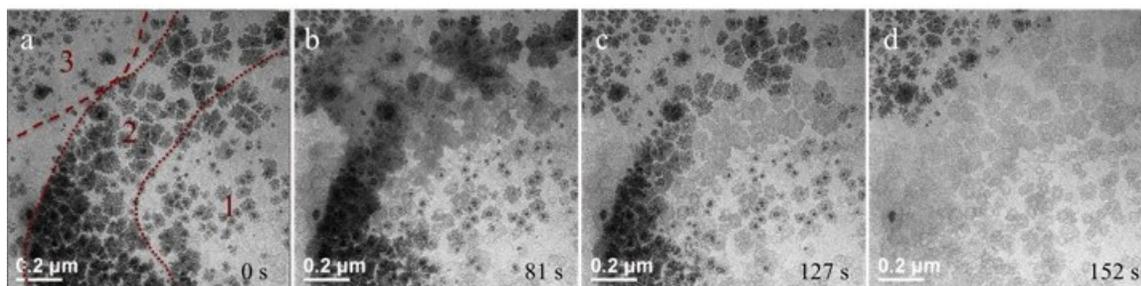


Figure S6. *In situ* TEM images of Pt deposits on 2D carbon electrode under potentiostatic polishing. (a), Before the polishing process, pristine deposits cover most areas of the electrode. Depending on the intensity of the deposits, electrode can be divided into three parts as marked by two dash lines. Area one has smallest deposits and area 2 has most intense ones. (b), Upon polishing, branch structured deposits chuted off and the gray contrast become dimmer. (c-d), Deposits vanished from main area.

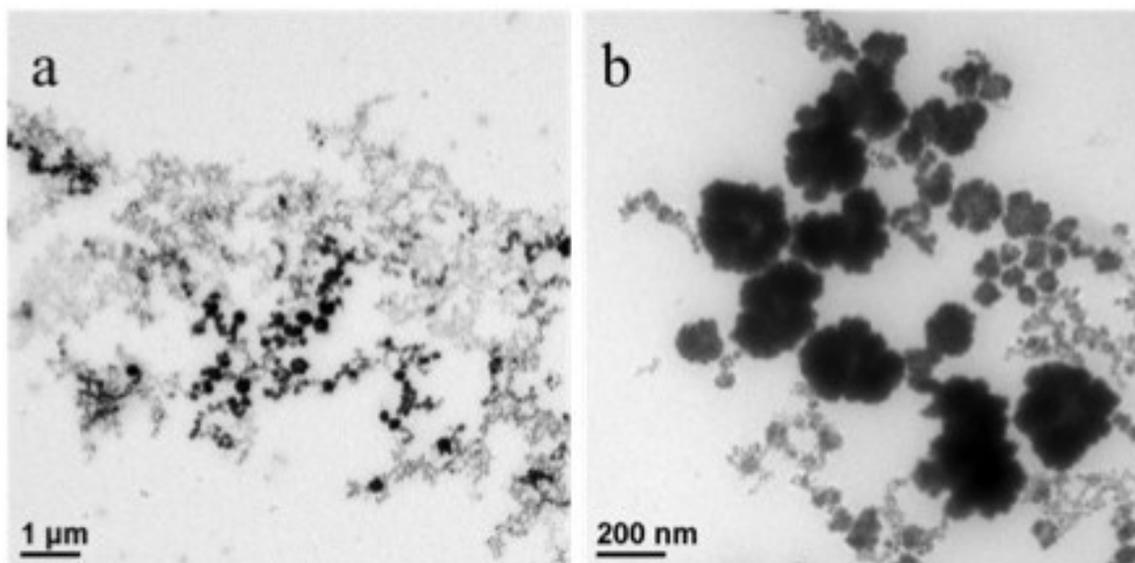


Figure S7. Surface of fractal nucleation islands serve as seed, atoms from electrolyte bath aggregate on top of it forming intense morphologies. Gray contrast tells the aggregated islands and newly formed intense deposit on top of it. (a), TEM image of the fractal islands of Pt deposits, dark dots being newly aggregated phase on top of the first layer. (b), TEM image of intense mesh islands as dark flakes forming on the first layer of aggregation fractal island.

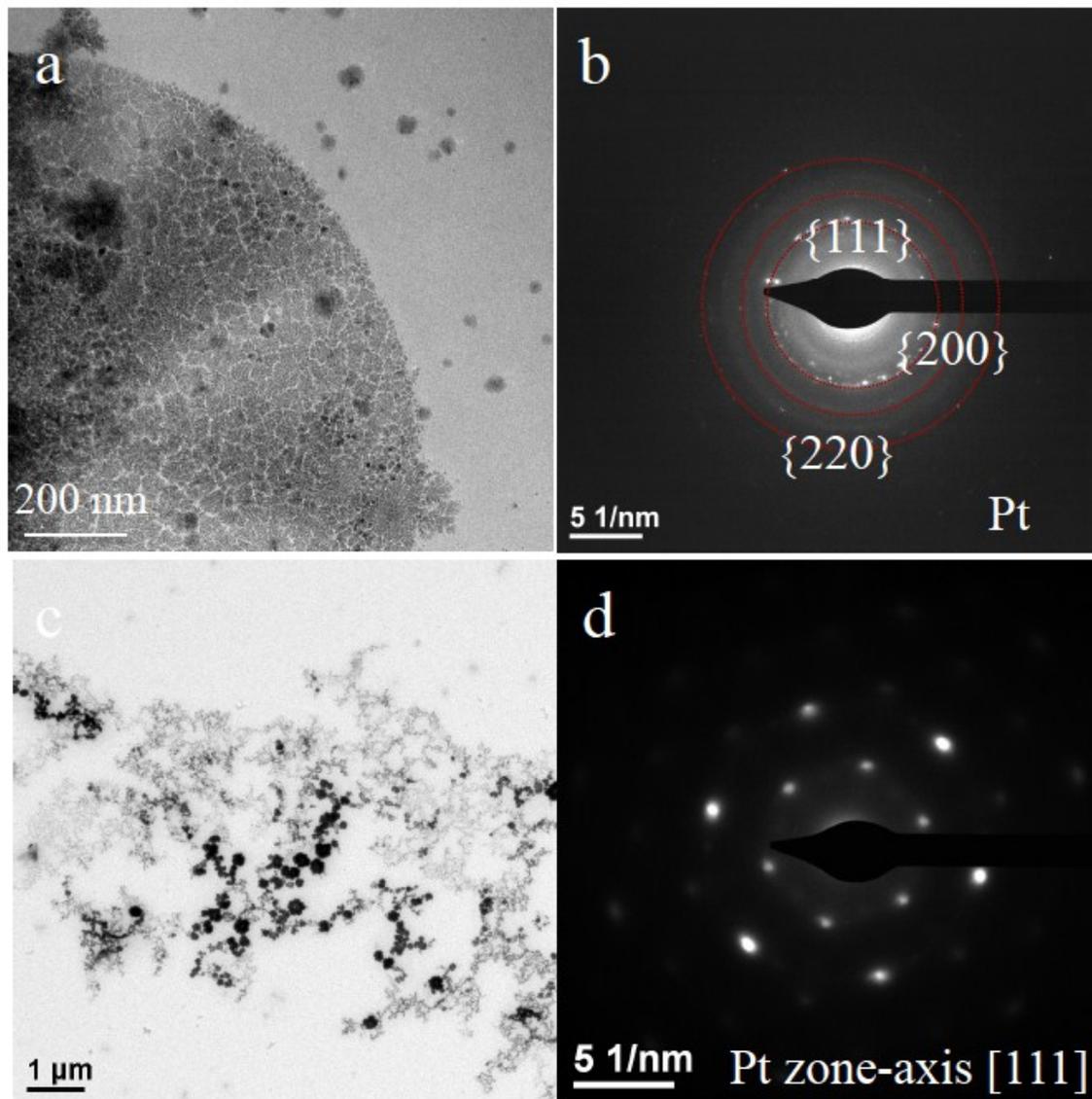


Figure S8. TEM bright field images of Pt electrodeposit and corresponding selected area diffraction patterns. (a, b) Ramified deposit with dense branching morphology shows a polycrystalline structure. (c, d) fractal deposit shows a single crystal phase when single particle was selected.

Supporting Movies

Movie S1. Record of Pt deposited from electrolyte bath onto glassy carbon electrode. In the viewing window, pristine glassy carbon is thick in the middle and thin in the other areas. Nucleation islands grow faster in thicker area than thin area.

Movie S2. Nucleation sites show up and grow bigger forming a uniform flat thin film on flat glassy carbon electrode. An impurity sitting in the electrolyte bath didn't show influence on the growth process.

Movie S3 and Movie S4. Records of nucleation sites show up and coalescence forming a fractal island.

Movie S5 and Movie S6. Concentration gradient driven growth of Pt deposition.

Movie S7. Polishing process of Pt deposits.