**Title:** Hierarchical Photonic Pigments via the Confined Self-Assembly of Bottlebrush Block Copolymers

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**Abstract:** Hierarchical, structurally colored materials offer a wide variety of visual effects that cannot be achieved with standard pigments or dyes. However, their fabrication requires simultaneous control over multiple length-scales. Here we introduce a new paradigm for the fabrication of hierarchical photonic pigments via the confined self-assembly of bottlebrush block copolymers within emulsified microdroplets. The bottlebrush block copolymer self-assembles into highly ordered concentric lamellae, giving rise to a near perfect photonic multi-layer in the solid-state, with reflectivity up to 100%. The reflected color can be readily tuned across the whole visible spectrum by either altering the molecular weight or by blending the bottlebrush block copolymers. Furthermore, the developed photonic pigments are responsive, with a selective and reversible color change observed upon swelling in different solvents. Our system is particularly suited for the scalable production of photonic pigments, arising from their rapid self-assembly mechanism and size-independent color.

**Summary of available data**

Supporting data for the article titled "Hierarchical Photonic Pigments via the Confined Self-Assembly of Bottlebrush Block Copolymers". The article was accepted for publication in 2019 in the journal "ACS Nano". Electronic supporting Information is available from the publisher (ACS). The data is provided within a structured set of folders compressed in zip, each correlating to a specific figure in the article. Spreadsheets are provided as `.xlsx`; images as `.bmp`, `.jpeg`, `.tiff` and `.png`; and NMR spectra are provided in `.fid` file format.

Nb. The individual tick of the scale bar images corresponds to a spacing of 10 µm.