Title: Aqueous Interfacial Gels Assembled from Small Molecule Supramolecular Polymers

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**Abstract:** The self-assembly of a stimuli-responsive aqueous supramolecular hyperbranched polymer from small molecules is reported. This system is composed of ditopic and tritopic guest-functionalised molecules that are able to form heteroternary supramolecular complexes with the macrocyclic host cucurbit[8]uril (CB[8]). We demonstrate that the supramolecular hyperbranched polymer formed is responsive to both photo- and chemical stimuli, exhibiting reversibility. Furthermore, this system is shown to assemble at liquid-liquid interfaces, which upon gelation, is observable on the micrometre scale. This self-healing supramolecular network can act as a soft matter barrier for aqueous microdroplets, inhibiting their coalescence.

# Summary of available data

Original or unprocessed data is provided in support of the article "Aqueous Interfacial Gels Assembled from Small Molecule Supramolecular Polymers". The data is structured into four folders, each correlating to a specific data type presented in the published article.

## Folder 1: Interfacial Tension measurements

Interfacial tension measurements are included as database files, and *txt* files including the relevant data. Images are also included as *jpg* format.

## Folder 2: Microscope & Photo Images

The optical microscopy images (.*png*) used in Figures 6, 7 and the ESI of the manuscript are provided with a scale bar included for reference.

## Folder 3: NMR Spectroscopy

<sup>1</sup>H NMR data files have been included as used throughout the manuscript and ESI.

#### Folder 4: UV-vis Spectroscopy

UV-vis data has been included as plotted in the ESI in *csv* format.