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S U P P O R T I N G I N F O R M A T I O N

On The Way to Biomimetic Dye Aggregate Solar Cells

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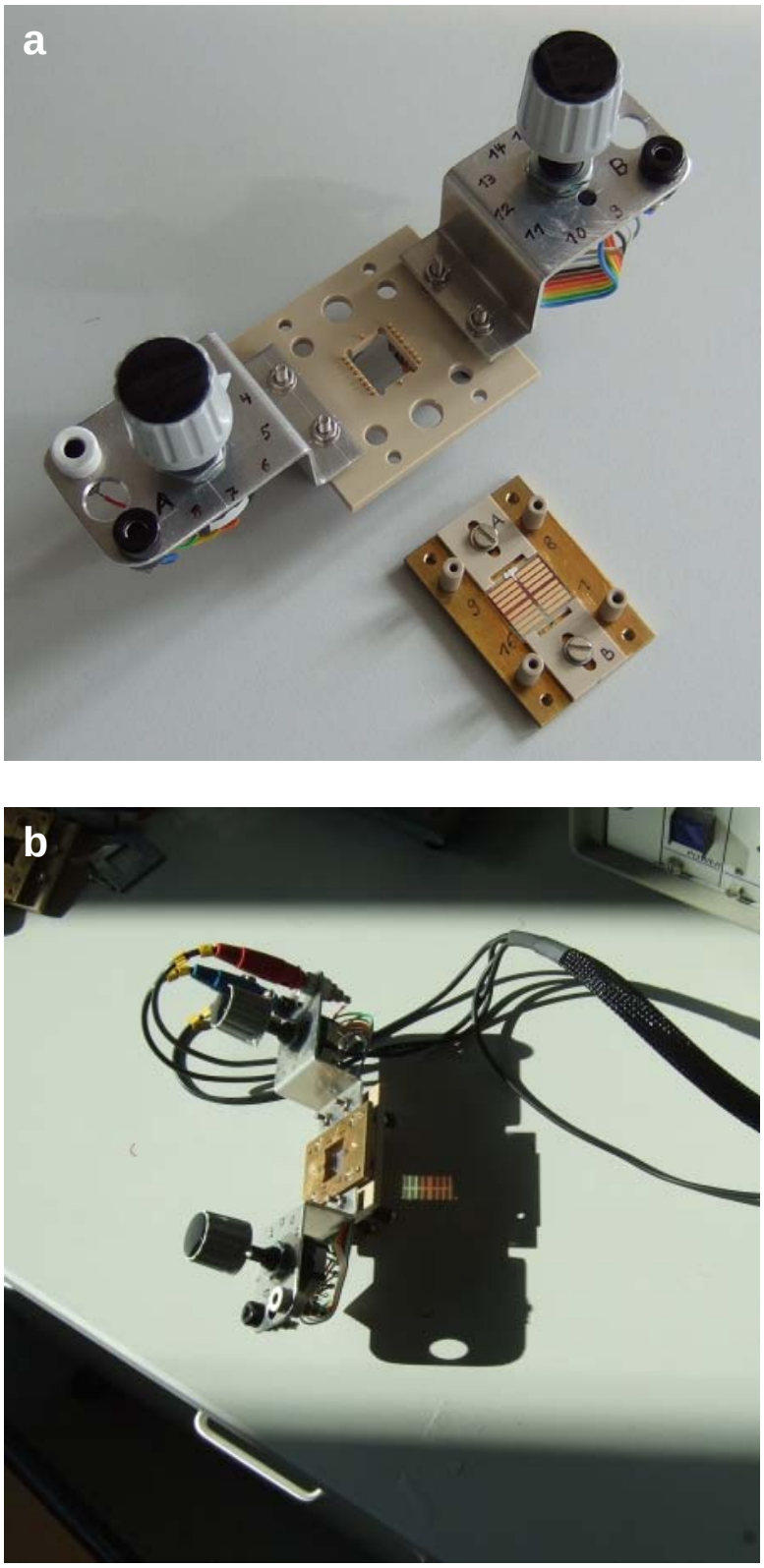

Figure S1. Pictures of the home built mounting (contacting) device for the I-V measurements of ss-DSSCs and DASCs, a) before contacting the reference ss-DSSC partly coated with a TiO_2 layer of 13 nm sized particles and sensitized by the ruthenium dye N719, b) ready contacted and illuminated from the FTO side by natural sunlight.

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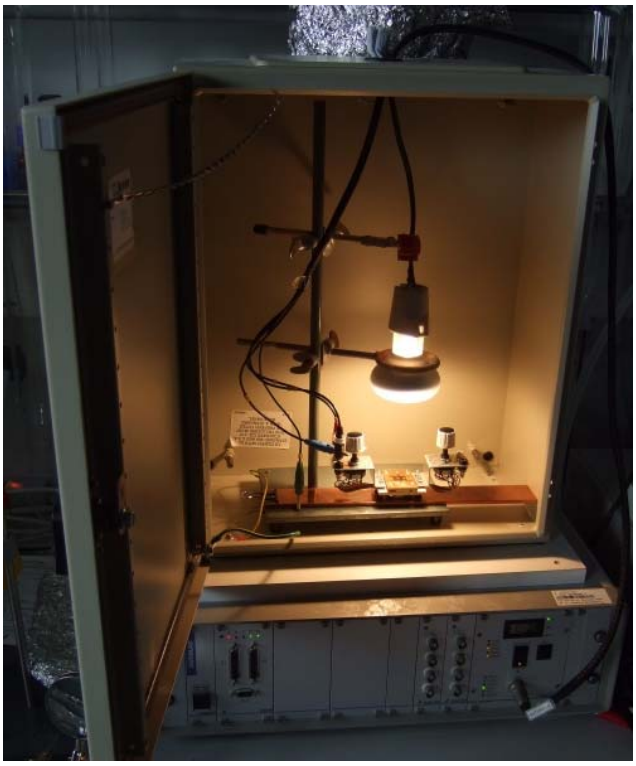

Figure S2. Setup for measuring the I-V characteristics under our standard indoor illumination conditions with a 60 W incandescent lamp (having a reflecting backside coating) from 10 cm distance.

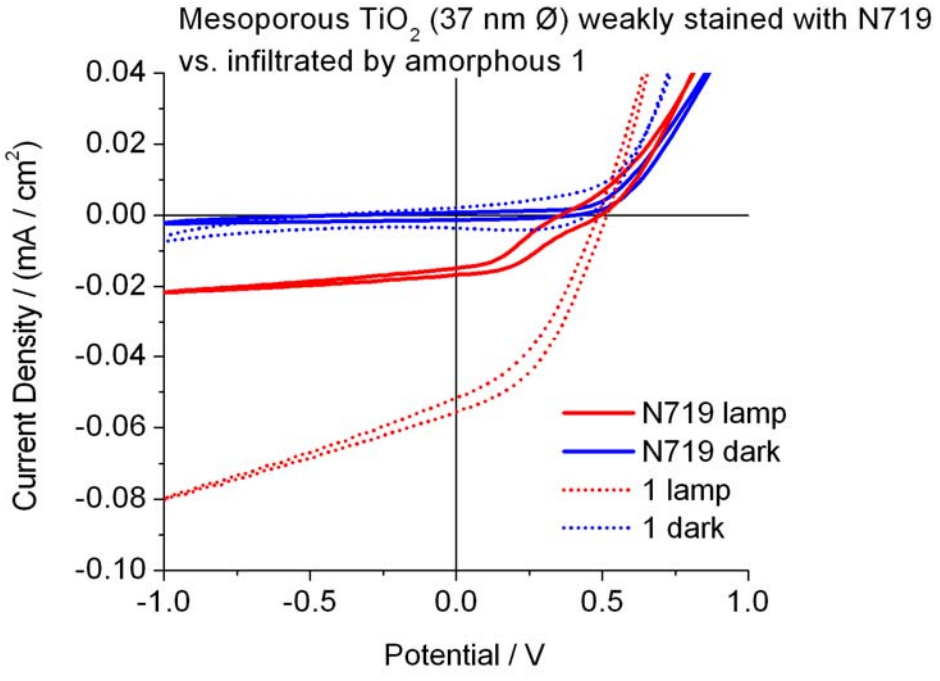

Figure S3. Performance comparison of a weakly stained N719-reference ss-DSSC and a DASC, based on an identically prepared mesoporous TiO_2 layer (from 37 nm TiO_2 particles) infiltrated with amorphous aggregates of **1** under our standard indoor illumination conditions. The hysteresis originated from a forward and backward sweep at 0.2 V/s. Summarized details: FTO / dense TiO_2 (ALD, 30 nm) / mesoporous TiO_2 (T37) / sintered 2 h at 450°C / (stained with N719) or (infiltrated with **1** by spraying from THF) / spiro-MeOTAD / 50 nm Au.

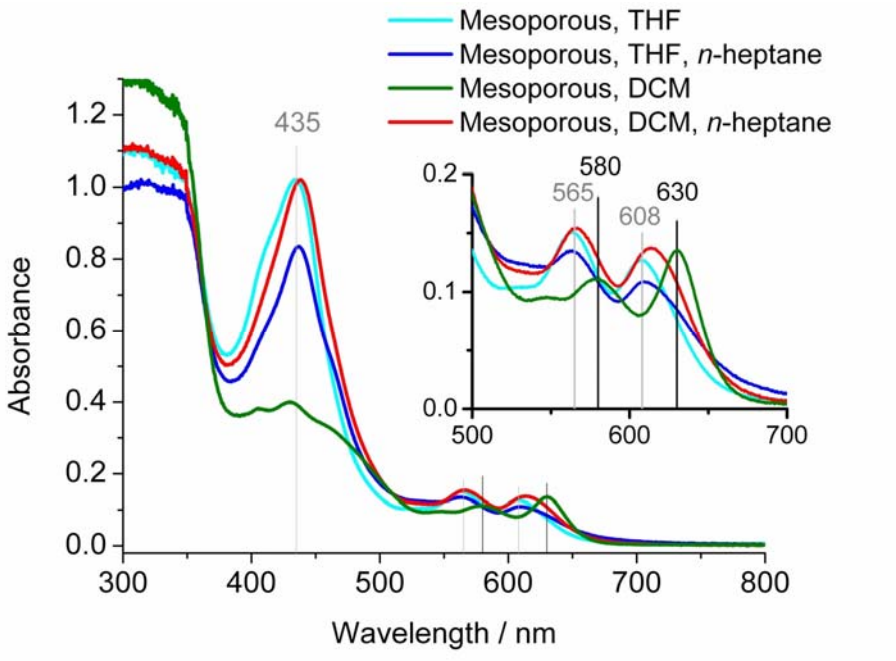

Figure S4. The absorption spectrum of a mesoporous TiO_2 layer (T37), infiltrated with **1** by spin-coating its 6 mM solution in anhydrous THF, revealed the presence of an amorphous phase of 1 (cyan trace). The following thermal treatment in *n*-heptane vapour at 110°C for 4 hours did not change the absorption spectrum significantly (blue trace). As reference J-aggregates of **1** have been spin-coated from a 6 mM dispersion in anhydrous dichloromethane onto a similar substrate (green trace). The following thermal treatment in *n*-heptane vapour surprisingly changed the absorption spectrum towards that of amorphous **1** (red trace). The inset shows a magnification of the Q-band region. The measurements were performed within an integrating sphere.

These measurements show that **1** does not self-assemble to J-aggregates within mesopores upon the treatment in *n*-heptane vapour and even existing J-aggregates disrupt and become amorphous by this treatment, most probably due to a diffusion of **1** from the surface into the mesopores. On flat or coarse-porous TiO_2 layers instead, the opposite behaviour was observed, namely the formation of J-aggregates upon the same treatment in the *n*-heptane vapour.