Supporting Information

for

Solvent-free sonochemistry: Sonochemical organic synthesis in the absence of a liquid medium

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Experimental part
1. **Experimental details**

All materials were purchased from Sigma-Aldrich UK with >98% purity. The particle size of the reagents was reduced by ball milling each reagent individually, in a 25 cm$^3$ stainless steel ball mill jar, with a 13.6 g ball bearing, at 25 Hz for 5 minutes (<500 µm) or 10 minutes (<200 µm) (employing a Retsch MM400 vibrational ball mill). The obtained powders were then sieved to obtain the desired particle size. Ultrasonic irradiation was carried out using a standard 35 kHz ultrasound bath. NMR spectroscopy was carried out with a Bruker Advance DPX 300 using CDCl$_3$ or DMSO-d$_6$ as solvent. Infrared spectroscopy was carried out using a Fourier Transform IR, Perkin Elmer Spectrum 1, with CsI beam splitter - 4000–200 cm$^{-1}$ using a pressed KBr disc. X-ray powder diffraction was carried out using a PANanalytical X'Pert Pro diffractometer with Cu Kα radiation.

2. **Synthesis of 1**

$O$-Vanillin of a particle size <200 µm (0.50 g, 3.29 mmol), 1,2-phenylenediamine of particle size <200 µm (0.36 g, 3.29 mmol) and anhydrous Na$_2$CO$_3$ (0.035 g, 0.329 mmol) were mixed in a 25 mL glass vial. The mixture was sonicated at 35 kHz for 10 minutes and then the ultrasonic bath was left to cool back to 25 °C (from ca. 30 °C). This process was repeated six times so that the sample was irradiated by ultrasound for 60 minutes. This produced a dark red solid. $^1$H NMR spectroscopy was carried out in CDCl$_3$.

![Figure S1: $^1$H NMR spectrum of 1 in CDCl$_3$/EtOD.](image-url)
Figure S2: PXRD patterns of 1 prepared from solution and ultrasonication.

Figure S3: IR spectra of 1 (green – sonication, blue – solution) with an imine stretching band at 1645 cm$^{-1}$ (with baseline corrected).
3. **Synthesis of 2**

Ninhydrin of a particle size <200 µm (0.50 g, 2.81 mmol) and dimedone of particle size < 200 µm (0.39 g, 2.81 mmol) were mixed in a 25 mL glass vial. The mixture was sonicated at 35 kHz for 10 minutes and then the ultrasonic bath was left to cool back to 25 °C (from ca. 30 °C). This process was repeated nine times so that the sample was irradiated by ultrasound for 90 minutes. This produced a pink solid. $^1$H NMR spectroscopy was carried out in DMSO-$d_6$.

**Figure S4**: $^1$H NMR spectrum of 2 in DMSO-$d_6$. 
Figure S5: PXRD patterns of 2 prepared from twin screw extrusion and ultrasonication.