

**Supporting Information**

**for**

**Mechanochemical Knoevenagel condensation**

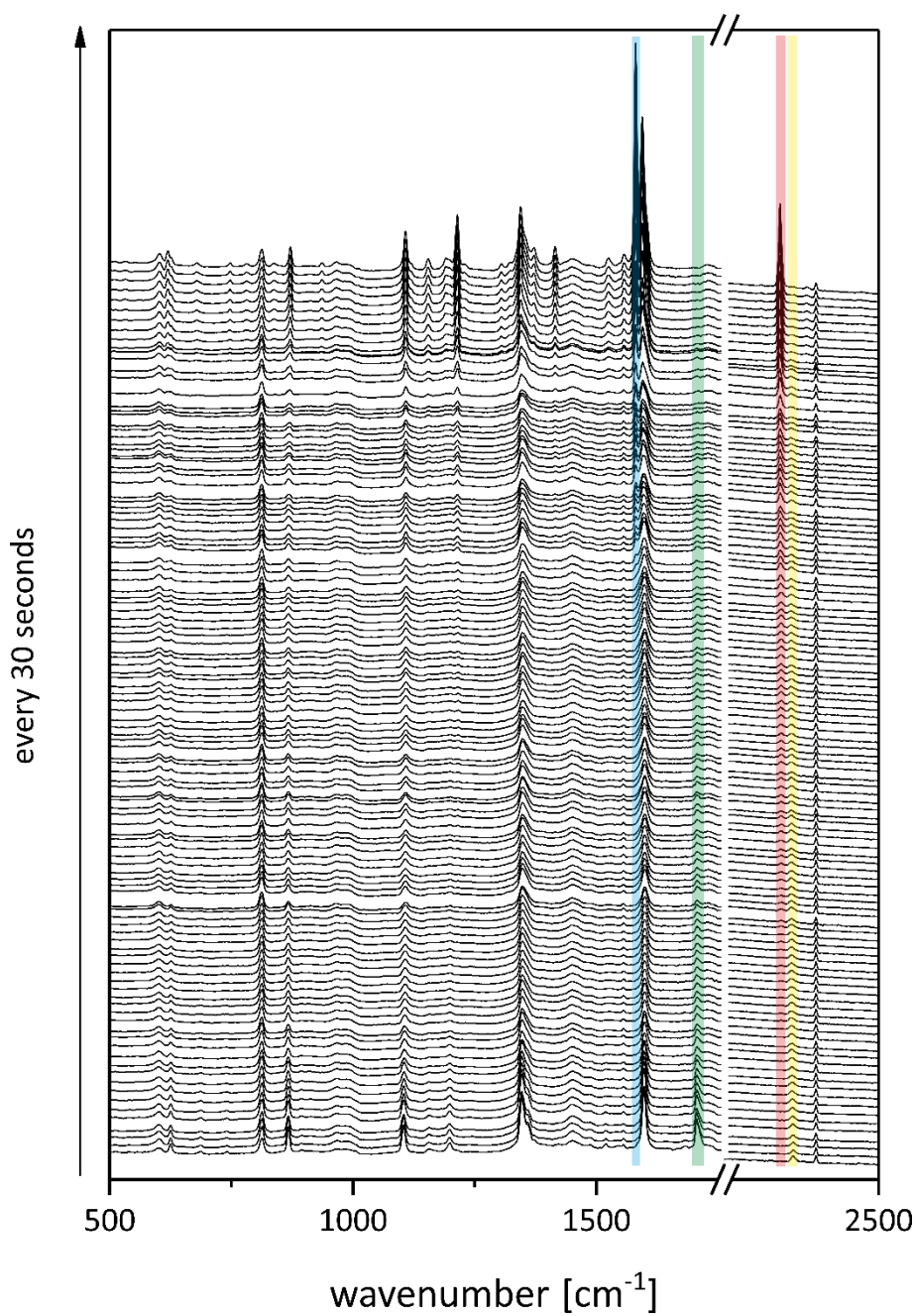
**investigated in situ**

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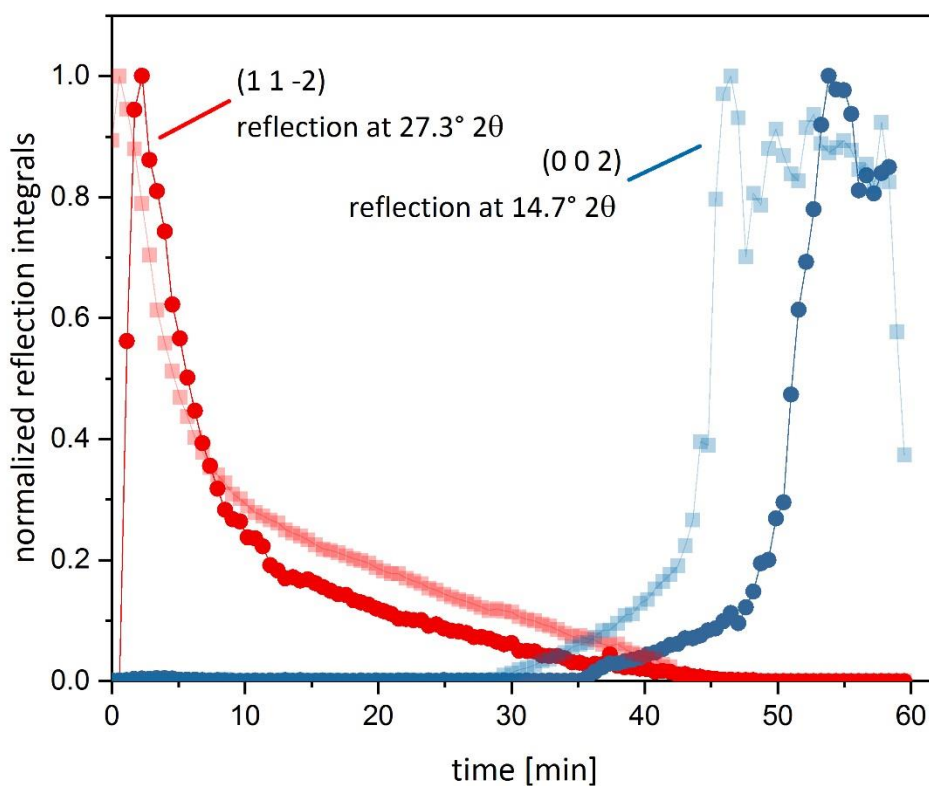
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**Raman spectra and XRPD data**



**Figure S1:** Time-resolved Raman spectra measured simultaneously. The progress of the reaction can be detected from the decreasing signal of the C=O-stretching band of **1** at  $1706\text{ cm}^{-1}$  (green) and the increasing signal of the C=C-stretching band at  $1581\text{ cm}^{-1}$  (blue). The band attributed to the C≡N-stretching shifts from  $2266\text{ cm}^{-1}$  (yellow) in **2** to  $2233\text{ cm}^{-1}$  (red) in the product.



**Figure S2:** Quantitative evaluation of the PXRD reflections intensities of selected reactant (red (11-2) reflection of **1**) and product reflections (blue (002) reflection of **3**). The PXRD data are representative measurements of repeated sets measured at two different beam times (intensive colour November 2016 and light colour August 2017). The data indicate a high reproducibility of the results. The course of the reaction can be divided in three steps. First a fast consumption of the reactant **1** is observed. After 11 minutes a decelerated consumption is observed. The formation of the product **3** starts after approx. 35 minutes and is characterized by a slow (until approx. 48 min.) and finally a very fast conversion.