## Supporting Information

## 1. NMR Spectra



Fig. S1: <sup>1</sup>H-NMR spectrum of the catalyst phase after the hydroformylation/aldol condensation reaction in water (top) and reference spectrum of sodium formate in water (bottom). Both measured in  $D_2O$ .

1.1. Reaction of NaOH to sodium formate in  $H_2O$  and PEG-200



**Fig. S2:** <sup>1</sup>H-NMR spectrum of the catalyst phase after the hydroformylation/aldol condensation reaction in water (top) and reference spectrum of sodium formate in PEG-200 (bottom). Both measured in  $D_2O$ .



Fig. S3: <sup>1</sup>H-NMR (top) and <sup>13</sup>C-NMR spectrum (bottom) of 2-butyloct-2-enal in CDCl<sub>3</sub>. Ca. 98% E-isomer.

## 1.2. Isolated 2-butyloct-2-enal (C<sub>12</sub>-Aldol)



## 2. Recycling Experiment with L-tryptophan as aldol catalyst

**Fig. S4**: Recycling experiment of the hydroformylation/aldol condensation of 1-pentene in PEG-200 with *L*-tryptophan. Conditions: Pentene (8.2 mmol, 0.9 mL), PEG-200 (0.4 mL), [Rh(acac)(CO)<sub>2</sub>] (0.1 mol%), sulfoXantphos (0.2 mol%), tryptophan (5 mol%), H<sub>2</sub>/CO (25 bar each), 100 °C, 16 h. Side products include pentane and alcohols.