Supporting Information for "Chemical Reaction-Induced Multimolecular Polarization (CRIMP) of Magnetic Resonance Imaging Agents."

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## **Supplementary Figures**



Figure S1: Schematic diagram of the dissolution DNP-MR setup.



**Figure S2:** Microwave frequency dependence of nuclear spin polarization levels of  $1,2^{-13}C_2$ -pyruvic acid for the OX063 radical.



**Figure S3:** Fit of the signal intensities of hyperpolarized  $H^{13}CO_3^-$  (red circle) and  $^{13}CO_2$  (blue triangle) with single exponential fit function. The pH value (black circle) was calculated employing Henderson-Hasselbalch equation at a given time point. pH values were determined using a calibrated AB15 Accumet Fischer Scientific pH meter with a micro pH electrode (13-620-95).



**Figure S4:** Intensity ratio between hyperpolarized  $H^{13}CO_3^-$  and  $^{13}CO_2$  resonances at two different pH values. pH values were determined using a calibrated AB15 Accumet Fischer Scientific pH meter with a micro pH electrode (13-620-95).