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Time-resolved X-Ray Absorption Spectroscopy of a Cobalt-Based Hydrogen Evolution System for Artificial Photosynthesis DOOSHAYE MOONSHIRAM, Argonne National Laborary, CAROLINA GIMBERT, Institut Català d'Investigació Química, CARL LEHMANN, STEPHEN SOUTHWORTH, Argonne National Laboratory, ANTONI LLOBET, Institut Català d'Investigació Química, ARGONNE NATIONAL LABORATORY TEAM, INSTITUT CATALÀ D'INVESTIGACIÓ QUíMICA COLLABORATION — Production of cost-effective hydrogen gas through solar power is an important challenge of the Department of Energy among other global industry initiatives. In natural photosynthesis, the oxygen evolving complex(OEC) can carry out four-electron water splitting to hydrogen with an efficiency of around 60%. Although, much progress has been carried out in determining mechanistic pathways of the OEC, biomimetic approaches have not duplicated Nature's efficiency in function. Over the past years, we have witnessed progress in developments of light harvesting modules, so called chromophore/catalytic assemblies. In spite of reportedly high catalytic activity of these systems, quantum yields of hydrogen production are below 40 % when using monochromatic light. Proper understanding of kinetics and bond making/breaking steps has to be achieved to improve efficiency of hydrogen evolution systems. This project shows the timing implementation of ultrafast X-ray absorption spectroscopy to visualize in "real time" the photo-induced kinetics accompanying a sequence of redox reactions in a cobalt-based molecular photocatalytic system. Formation of a Co(I) species followed by a Co(III) hydride species all the way towards hydrogen evolution is shown through time-resolved XANES.

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