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Data Synchronization in Operando Gas and Heating TEM

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Operando gas and heating TEM is attracting more and more research interests in the catalysis community for its capabilities in correlating the materials' gas environment, atomic structure and reaction products at high spatial and temporal resolutions [1-4]. One important, while could be ignored is the time delays between different parameter measurement locations. Simultaneously measured data don't naturally mean they are synchronized. In a typical Operando TEM set-up, as shown in Figure 1(a), the gas needs to travel from a Gas Supply System (GSS) into the TEM, and then from the TEM to the Mass Spectrometer (MS). Figure 1(b) shows that a user set gas composition change in the GSS will show changes in the MS after 79.1 s. The intrinsic time delays that exist among parameter measurement locations need to be calibrated for valid correlations. In this work, we will present a data synchronization method in operando gas and heating TEM based on time delay calibrations enabled by the nano-calorimetry localized on TEM Nano-Reactors.

The experimental operando TEM set-up used the DENSsolutions Climate Gas Supply System (GSS), Climate in-situ TEM gas and heating sample holder in a Thermo Scientific Themis ETEM and Climate in-situ gas analyzer. The gases H_2, CH_4, O_2, CO_2 of 99.999% or higher purity are used as purchased. An empty Nano-Reactor, without any samples on it, was used for systematic investigations.

With detailed understanding of the experimental set-ups, we investigated these parameters' influence on time delay: gas flow rates (F_{NR}), gas composition and gas pressure (P_{NR}) inside the Nano-Reactor, and the tubing length (L) in connecting the hardware. We discover that the time delay depends on the F_{NR} and P_{NR}, and has little dependence on the gas type. Based on the experimental observations and the theoretical derivations, a functional relationship has been derived, which can be written as

$$t = \frac{eP_{NR}}{a+bF_{NR}+cF_{NR}{}^2+dF_{NR}{}^3} + f \quad (1)$$

A glimpse of the experimental data and the fitted equation is shown in Figure 2. More results will be presented on site.

Afterwards, open source codes have been developed to achieve automated data synchronization on-line and off-line. Finally, we propose a general protocol to perform automatic time delay calibration for general operando TEM set-ups. The current research results and conclusions are expected to speed up the valid data synchronization and ensure more reliable information correlations in operando gas and heating TEM.

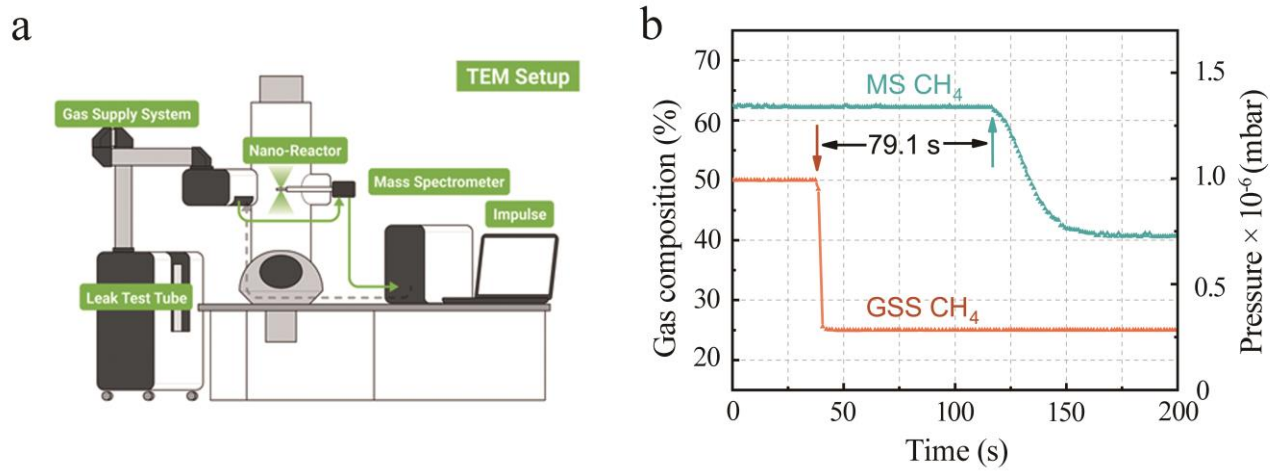

Figure 1. (a) A schematic view of a gas cell based operando gas and heating TEM setup; (b) Illustration of time-delay between GSS and MS.

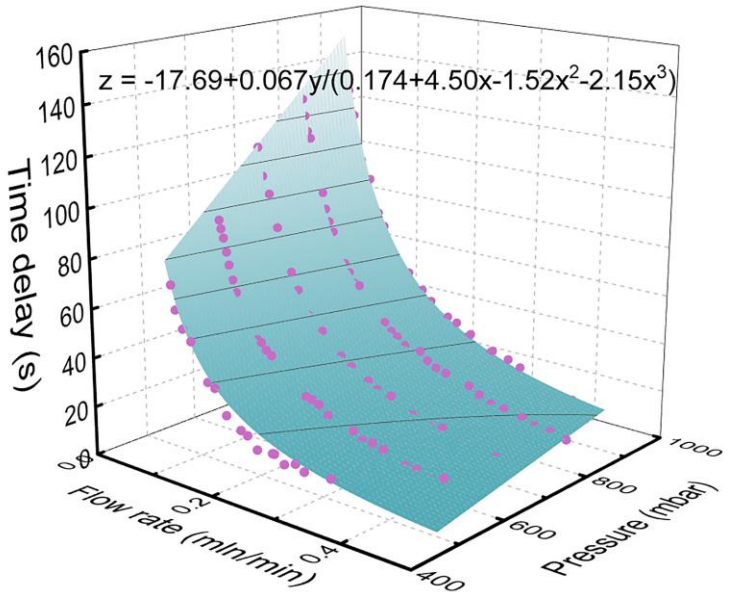

Figure 2. A fitted function relationship of time delay to F_{NR} and P_{NR}.

References:
[1] Hansen, P.L., Wagner, J.B., Controlled Atmosphere Transmission Electron Microscopy. 2016, Cham: Springer International Publishing.
[2] Vendelbo, S.B., et al., Visualization of oscillatory behaviour of Pt nanoparticles catalysing CO oxidation. Nature Materials, 2014. 13(9): p. 884-890.
[3] Tan, S.F., et al., Real-Time Imaging of Nanoscale Redox Reactions over Bimetallic Nanoparticles. Advanced Functional Materials, 2019. 29(37).
[4] Plodinec, M., et al., Insights into Chemical Dynamics and Their Impact on the Reactivity of Pt Nanoparticles during CO Oxidation by Operando TEM. Acs Catalysis, 2020. 10(5): p. 3183-3193.

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