doi:10.1017/S1431927617005244
Microsc. Microanal. 23 (Suppl 1), 2017

The Effect of Gas on Image Quality and Resolution in *In situ* Scanning Transmission Electron Microscopy

Yuanyuan Zhu[1], Nigel D. Browning[1, 2]

[1.] Physical & Computational Sciences Directorate, Pacific Northwest National Laboratory, Richland, WA 99352, USA
[2.] Department of Materials Science and Engineering, University of Washington, Seattle, WA 98195, USA

In situ environmental (scanning) transmission electron microscopy in gaseous environment has opened up new opportunities for studying the dynamics of gas-solid reactions in import applications such as nanomaterials growth and heterogeneous catalysis. Although environmental ADF-STEM (ESTEM mode) has began to demonstrate advantages in particular for offering directly interpretable atomic dynamics of complex oxide catalysts[1], the role of gases in determining the quality of environmental images has so far only been discussed for the HR-ETEM imaging mode[2][3]. For quantitative ESTEM analysis, it is therefore crucial to understand how the gas molecules affect the scattering of fast electrons contributing to the image formation.

In this work, we conducted the systematic experimental evaluation on the signal-to-noise ratio (SNR), contrast and resolution of ESTEM images under the influence of different gases at various pressures, in a differentially pumped (DP) gas cell. The DP-ESTEM assessment was carried out on a dedicated Titan ETEM operated at 300kV, with convergence $\alpha = 9.9$ mrad and collection $\beta = 20.9 \sim 71$ mrad, and a low electron dose-rate of 4.79×10^1 e/Å^{2}s for atomic imaging. Starting with a standard lacy carbon, in Figure 1, we found that unlike the ETEM, there is no intensity loss in the ESTEM mode as both of the background intensity I_{gas} and the solid sample intensity I_{carbon} increase with the gas pressure. The apparent fade-out contrast at high pressures is mainly due to the low atomic number of the carbon specimen. For the complex oxide MoVNbTeOx M1 catalyst (Z ~16.8), sharp contrast was maintained up to 10 mbar in N_2 (Figure 2a). Importantly, our measurement based on the SNR defined by studies on the ETEM mode[4] pointed out that the ESTEM background SNR_{gas} and the SNR_{carbon} were excellent in gas and remained high (> 400) in 10 mbar Ar. These observed uncompromised intensity and the extraordinary SNR_{gas} in the ESTEM images can be explained by the nature of the electrons this mode collects for image formation (Figure 1d). In particular, the ESTEM imaging mode is largely free from the inelastic scattering effects induced by the presence of gases and hence retains an excellent SNR.

For the atomically resolved ESTEM, our tests on the M1 catalyst have shown that the specimen SNR_{M1} and resolution depend on the nature and the pressure of the gas, and on the accumulated electron dose. As shown in Figure 2, STEM probe broadening based on multiple-scattering theory of equivalent solid foil thicknesses of different types of gases was calculated, and the it matches well with the experimental observations at a relatively high dose of ~ 10^3 e/Å^2. Similar discussions on the theoretical obtainable resolution can be extended to membrane-windowed ESTEM gas cell to provide guidance for atomic *situ* ESTEM investigations. In sum, the influence of gases on the ESTEM is likely two-fold - the pre-specimen gas causes probe broadening and determines the obtainable ESTEM resolution, and the post-specimen gas leads to imaging signal diffusion and thus electron dose dependency. The property that both of the SNR and resolution of ESTEM images increase along with the accumulation electron dose is beneficial for achieving high-sensitivity, and at the same time, high-resolution ESTEM observations.

References:

[1] Y Zhu, PV Sushko, D Melzer, E Jensen, L Kovarik, C Ophus, M Sanchez-Sanchez, J Lercher, ND Browning, (2017), In review.
[2] JR Jinschek, S Helveg, Micron **43**(2012), 1156
[3] M Suzuki, T Yaguchi, XF Zhang, Microscopy **62**(2013), 437
[4] This research is part of the Chemical Imaging Initiative conducted under the Laboratory Directed Research and Development Program at Pacific Northwest National Laboratory (PNNL). PNNL, a multiprogram national laboratory, is operated by Battelle for the DOE under Contract DE-AC05-76RLO1830.

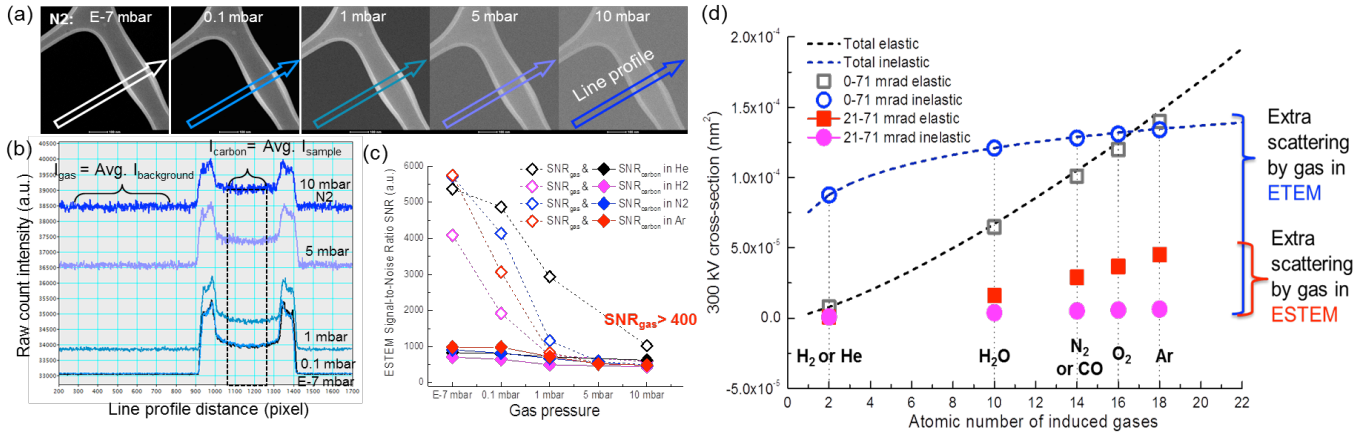

Figure 1. a) Gas pressure series, b) line profiles and c) SNR_{gas} and SNR_{carbon} of ESTEM images of gas background and amorphous lacy carbon. d) Theoretical comparison of the cross sections of elastically and inelastically scattered electrons between the ETEM and the ESTEM imaging mode.

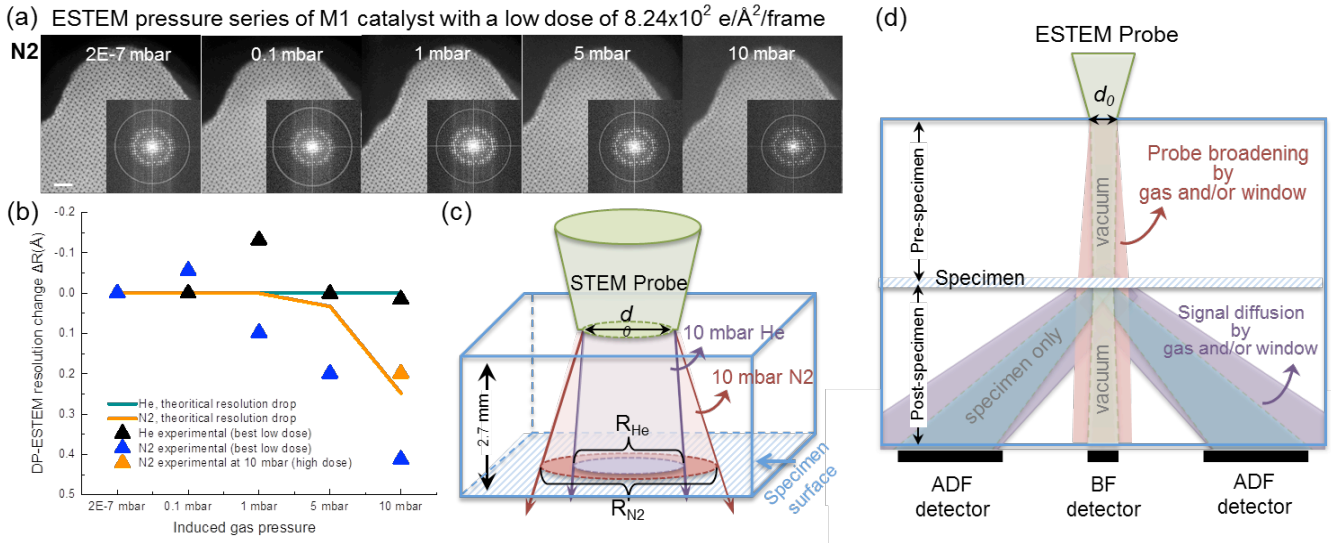

Figure 2. a) Gas pressure series of atomic ESTEM images of complex oxides M1 catalyst. Scale bar is 5 nm. b) Experimental and theoretical DP-ESTEM resolution over gas type and pressure. Note that ESTEM resolution is electron-dose dependent. c) Pre-specimen gas introduced STEM probe broadening. d) Summary of the pre- and post-specimen gas effects on ESTEM probe and signal.

https://doi.org/10.1017/S1431927617005244 Published online by Cambridge University Press