

***In situ* gas-phase monitoring of pyrolysis and *ex situ* assessment of carbon contamination in MOCVD grown MoS₂ films**

Christian M. Schäfer

J. Caicedo Roque, G. Sauthier, E. Del Corro, J. R. Sperling, J. Santiso, J. A. Garrido

Catalan Institute of Nanoscience & Nanotechnology (ICN2), CSIC and The Barcelona Institute of Science and Technology, Bellaterra (Barcelona), Spain

christian.schafer@icn2.cat

Metal-organic vapor deposition (MOCVD) is the method of choice for scalable and controllable film deposition of transition metal dichalcogenides (TMDs) on large areas [1]. However, there are still open questions about the optimal synthesis conditions depending on the used precursor chemistry. While carbon-free chalcogen hydrides have been demonstrated as viable precursors, safety concerns given their high toxicity have motivated further research into their less harmful organic counterparts. However, the use of such organic chalcogen precursors can introduce carbon (C) contamination through C-containing pyrolysis products, impeding the growth of high quality, two-dimensional TMD layers. [2][3][4] Therefore, a better understanding of the pyrolysis mechanism leading to C contamination and its effect on TMD growth and film properties is necessary.

Here, we employ *in situ* gas-phase analysis using a mass spectrometer together with *ex situ* film characterization of MoS₂ grown by MOCVD in a hot-wall reactor from Mo(CO)₆ and the organic chalcogen precursor (CH₃CH₂)₂S (diethyl sulphide, DES). By systematic study of the gas-phase composition of single and multi-source DES, DES:Mo(CO)₆ and DES:Mo(CO)₆:H₂ mixtures and varying temperature, we observed that DES pyrolysis by an unimolecular dissociation pathway involves ethyl radical/ethylene production. We have investigated the effect of the DES pyrolysis on the MoS₂ film by monitoring the C contamination level via semi-quantitative post-growth characterization, including scanning electron microscopy, Raman, photoluminescence (PL) and X-ray photoelectron spectroscopy.

From these studies we conclude: (1) Thermal DES decomposition acts as the source of C contamination for growth temperatures above pyrolysis onset, typically around 600°C; (2) Unintended formation of graphitic carbon and C_xS_y surface compounds hinder lateral MoS₂ grain growth resulting in interrupted MoS₂ film morphology and non-ideal stoichiometric composition; (3) PL is quenched gradually with increasing C impurities suggesting charge transfer doping; and (4) Reductive H₂ gas effectively suppresses DES pyrolysis, thus lowering C contamination, even at growth temperatures above 700°C.

This work contributes to the growing body of research investigating the use of low-cost, low-toxicity organic chalcogen precursors in gas-source MOCVD, aiming at the development of contamination-free TMD films for high-performance devices.

Acknowledgement

This work was funded by the EU Horizon 2020 programme under Grant Agreement No. 732032 (BrainCom). The ICN2 team was supported by the Severo Ochoa program from Spanish MINECO (Grant No. SEV-2013-0295), and by the CERCA Programme/Generalitat de Catalunya.

References

- [1] Kang K., et al., Nature, Vol. 520: 7549 (2015) 656-660
- [2] Schäfer C. M., et al., Graphene Conference (2019), Rome, poster contribution
- [3] Choudhury T. H., et. al., Cryst. Growth Des., Vol. 18: 8 (2018) 4357-4364
- [4] Zhang, X., et. al., Journ. Elect. Mat., 45: 12 (2016) 6273-6279