Electrodeposition of Transition Metal Dichalcogenide Thin-films from Non-aqueous Solvents

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Transition metal dichalcogenides (TMDCs) are an interesting group of 2D materials characterised with a layered structure analogous to graphene and they possess unique electronic and optical properties, especially when in the few- and mono-layer form. Developing scalable techniques for depositing TMDCs is a major challenge which needs to be overcome to fabricate functional devices with these materials. Electrodeposition is an industrially relevant technique that has some key advantages over other conventional deposition methods. It is a low cost and easily scalable technique and could be used for obtaining complex nanoscale features and for depositing over topologically demanding surfaces. Even though the electrodeposition of MoS_2 has been achieved both in aqueous and non-aqueous electrolytes, not much progress has been made in the deposition of other TMDC materials. Tungsten based TMDCs such as WS_2 and WSe_2 are shown to be very promising materials in different applications, however electrodepositing them remains extremely challenging. One of the major obstacles here is developing electrochemically active precursors which are compatible with the electrolyte system and able to deliver both the tungsten and chalcogens to the electrolyte. Controlling and optimizing the deposition process to obtain few- and mono-layer TMDCs is another challenge. In addition, the choice of substrates for deposition is very important, especially for direct growth of ultra-thin TMDCs. Electrodeposition, being a bottom-up deposition method, would benefit from an atomically thin and smooth substrate such as graphene for depositing few- and mono-layer TMDCs.

Here we present non-aqueous electroplating as a scalable alternative technique for tungsten-based TMDC deposition, with WS_2 and WSe_2 as examples. Tailored single source precursors were developed to use in non-aqueous electrolytes. WS_2 was electrodeposited from dichloromethane (CH_2Cl_2) using the $[NEt_4]_2[WS_2Cl_4]$ precursor. WSe_2 was then electrodeposited from acetonitrile (CH_3CN) electrolyte using $[WSeCl_4]$ as the precursor. Electrochemical quartz crystal microbalance (EQCM) studies were performed to optimize the deposition process and to probe the mechanism of precursor electrochemistry. Electrochemical deposition parameters were then carefully adjusted to obtain few- and mono-layer TMDCs. Patterned graphene electrodes were used as an atomically thin and smooth platform for the deposition of few- and mono-layer WS_2. Few-layer TMDC films obtained on graphene were found to be much smoother than films deposited on other standard substrates such as titanium nitride (TiN) or Pt. These TMDC/graphene structures gave interesting 2D heterostructures which are technologically important for different applications.

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