Advanced energy storage devices and carriers such as hydrogen, supercapacitors, fuel cells and batteries has been regarded as the clean, efficient and renewable energy supplier to meet the future world energy demand. Therefore, searching for good electrode materials is of importance for the implementation of sustainable energy devices and energy carriers. Transition metal dichalcogenides (TMDs) as well as other layered materials have been used as the electrode materials in various electrochemical applications due to the intriguing optical, electrical and electrochemical properties. This thesis investigated a spectrum of TMDs and layered materials to reveal their electrochemical properties and their potential as the electrocatalysts toward hydrogen evolution reaction (HER). Layered post-transition metal chalcogenides like gallium chalcogenides and indium chalcogenides as well as the Group 5 TMDs have been comprehensively studied to reveal their fundamental electrochemical properties and electrocatalytic activity. Furthermore, novel strategies including exfoliation of TMDs, phase engineering and TMD hybrids with conductive graphene have been utilized to tune and improve the electrocatalytic activity toward HER. Moreover, electrochemical concept of bipolar electrochemistry was adopted to exfoliate and downsize the layered materials for electrochemical applications with improved HER performance.