Montpellier, April 30th 2019

Object: Report on the dissertation "Theoretical Investigation of Novel Two-dimensional Materials with Application Potential" by M.Sc. Pengbo Lyu, Charles University, Faculty of Science.

The PhD project deals with a systematic computational exploration of the electronic and structural properties for a broad range of 2D-materials (COFs, several Transition metal-based materials, LDHs and Silicene) using advanced quantum-based calculations. I am very impressed by the massive work the candidate successfully achieved during his PhD. Besides an important fundamental understanding of many different properties (magnetism, catalysis activity, lithium ion battery) of the investigated systems at the microscopic scale, the candidate had the talent to make a correlation between these properties and the structural and electronic features of the materials to further guide the experimentalist towards refined materials with improved performances.
The thesis starts with a clear and concise introduction and statement of purpose. All chapters suitably start with a brief introduction and the contributions to the chapter and contain a summary of the main conclusions.

Chapter 1 presents a brief introduction of the 2D materials selected in the thesis and the main challenges that need to be addressed for all of them in terms of atomistic understanding of their structures and related properties. The chapter ends by an overview of the following chapters and the computational strategy (mostly GGA DFT) that was applied to tackle all the materials.

Chapter 2 describes the basic principles of the DFT methods as well as the details of the calculations that were performed to assess the electronic-based properties of the 2D-materials. The candidate also listed the programs used as well as the functional/basis set. It would have been interesting to have a discussion on section 2.10 about the comparison between the performances of the different functional PBE, PBE0 and HSE for the different targeted properties in the selected 2D-materials rather than only mentioning a general statement. More details about the 2S Ising model would have been also required rather than providing the link to a webpage (page 18).

Chapter 3 reports the whole results that have been collected on the diverse 2D-materials starting with the CFT-1, HPP and TzF COFs. The candidate first explored the stability of the corresponding system depending on the arrangement of the layers (helical, eclipsed, staggered,…)

Prof. Guillaume Maurin– Member of Institut Universitaire de France
Head of the group « Dynamics & Adsorption in Materials with Porosities », Institut de Chimie Moléculaire et des Matériaux de Montpellier UMR 5253
Université Montpellier – Place Eugène Bataillon – Bat. 15 –CC 1505
34095 Montpellier cedex 5 France
☎ +33467143307 – guillaume.maurin@univ-montp2.fr

that is affected by the consideration of the dispersion correction followed by a careful analysis of the calculated band gap and edge position for all the systems and a correlation with their geometric (interlayer arrangement/distance…) and chemical features (N content,….). The candidature further investigated this family of COFs in collaboration with experimentalists to explore their performances in terms of alkali ion battery which led to simulated specific capacity and open circuit voltage in good agreement with the corresponding experimental data. This section was followed by the computational exploration of the silicone and transition metal-based systems for Lithium-ion battery and CO catalytic oxidation. In particular the work performed on the Fe-$PtSe_2$ is very interesting with a full understanding of the catalytic reaction mechanism. The section 3.4 was devoted to the computational exploration of the magnetism properties of transition metal carbides. Although the results are well described, I would have appreciated more details on the Monte Carlo simulations employed to predict the Curie Temperature of these systems. Finally the chapter ends with the computational exploration of the LDHs in strong interplay with experimental data to characterize the structure of these materials as well as the mechanism of carbonate grafting.

Chapter 4 finally closes the manuscript with a brief summary of the main results achieved during the PhD. I would have expected the candidate to provide some perspectives of this work.

Overall, the academic English and formatting is of good quality throughout the manuscript and the scientific content is excellent as monitored by a rather impressive list of already published papers (12). For this reason, I give my strongest support for the defense of this dissertation that fulfills the requirements for awarding the degree of doctor.

Sincerely yours

Prof. Guillaume MAURIN

Prof. Guillaume Maurin– Member of Institut Universitaire de France
Head of the group « Dynamics & Adsorption in Materials with Porosities », Institut de Chimie Moléculaire et des Matériaux de Montpellier UMR 5253
Université Montpellier – Place Eugène Bataillon – Bat. 15 –CC 1505
34095 Montpellier cedex 5 France
☎ +33467143307 – guillaume.maurin@univ-montp2.fr