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Clermont-Ferrand, le 06/09/19

Report on the Habilitation Thesis of Fatima Hassouna

Dr Hassouna started her research work in France, successively at the university of Lille and at the university of Clermont-Ferrand. On the base of the addresses indicated in her publications, she joined then research teams in Luxembourg, the Public Research Centre Henri Tudor and the Luxembourg Institute of Science and Technology. Finally, she integrated the University of Chemistry and Technology of Prague as an assistant professor. Since obtaining her PhD in 2006, Dr Hassouna has acquired 13 years of research experience.

In her habilitation thesis, Dr Hassouna presents first her research activity dedicated to the design of high performance polymeric materials with tailored morphology and properties. In her work, eco-friendly methods are favoured, as water-borne systems and free-solvent processes. **This part gives the necessary elements to understand her current research activity and perspectives.** In the second part of the habilitation thesis, she lists 25 papers she has published in impacted journals, followed by a copy of these articles. This gives the reviewer the possibility to deepen the elements given in the general presentation and to have an overview of Dr Hassouna's global research activity.

The presentation starts with an introduction, followed by two main parts. The first part deals with a new solvent-free methodology for the preparation of super-tough materials by reactive-extrusion of high performance polylactide (PLA). The second part concerns three types of novel nanocomposite materials with tunable properties. Finally, Dr Hassouna presents new research developments and perspectives of her work.

In **chapter 1 - introduction** -, Dr Hassouna underlines the fact that polymers are resistant to degradation in environmental conditions and usually non-biodegradable, is both an advantage for their use properties and a disadvantage at the end of their life circle, when they remain for hundreds of years in nature. In order to reduce environmental impact, she explains that her research is based on the development of alternative methods of preparation, including the use of renewable resource instead of petrol, and of water-based or solvent-free processes. Environmental impact is taken into account in all the research activity presented.

Chapter 2 is dedicated to the design of high performance PLA and to the study of its thermo-mechanical recycling. This commercial polymer is compostable and synthesised with sustainable resources. In order to improve its mechanical properties, chemical modification via reactive grafting of different functionalized plasticizers is developed. Reactive plasticization of PLA is realized with **low molecular weight poly(ethylene glycol) (PEG) derivatives**. Two unsaturated methacryl functional reactive PEG were oligomerized and grafted onto PLA chains in presence of a free-radical initiator. The exploitation of the numerous analyses realized (Soxhlet extraction, FTIR, SEC, 1H NMR, DSC and DMTA, SEM, Peak Force AFM, tensile tests and notched Izod Impact) has permitted to draw a mechanism of reactive grafting. Analyses also demonstrated an improvement of the mechanical and thermal properties, and gave information about the microstructure of the obtained material. The degradation mechanism occurring during the thermo-mechanical recycling was also studied. **Cardanol and citrate derivatives** graftings were also investigated. Other studies on PLA-based materials were also carried out and are not reported in the general presentation. With 12 papers on the design of high performance PLA materials corresponding to different goals and implemented strategies, Dr Hassouna is a specialist of this polymer and its derivatives.

Chapter 3 is devoted to the fabrication of three different nanocomposites. The first nanocomposite developed concerns the reinforcement of a polymer by biobased nanofillers. The nanofillers chosen are cellulose nanocrystals (CNC). CNC present all the required properties for the preparation of the nanocomposite, except their polar nature that requires the use of a compatibilizing strategy with hydrophobic polymeric matrix. The methodology developed consists in the incorporation of a CNC aqueous suspension into a polymeric latex suspension. The material obtained is a combination of a hard core and soft shell matrix (CS15) with a CNC reinforcing agent and polyethylene glycol (PEG) as plasticizer. Adjusting the ratios of these three components permitted to tailor the desired properties.

The second nanocomposite, made of polyaniline (PANI) and reduced graphene oxide, find applications in the domain of energy storage devices. Loading PANI with reduced graphene oxide (RCO) is expected to improve its cycle stability and its degradation at relatively high potentials. An interesting synthesis of these nanocomposites by "ex-situ reduction" was developed and compared to an "in-situ reduction" way. A thorough study of the properties of the obtained materials including cycling voltammetry was carried out. Unfortunately, the data suggested that no protection was provided by RGO. This work is actually submitted to publication.

The third nanocomposite has been elaborated in order to capture carbon dioxide by the means of nitrogen groups incorporated in a porous carbon matrix (PANI). This new approach is developed with the ambition of creating micropores, mesopores and macropores – a 3D hierarchically porous material - for high CO_2 adsorption and facilitated CO_2 diffusion. Spherical polystyrene latex nanoparticles were chosen as building blocks and removed after the synthesis of the PANI skeleton in presence of a crosslinker. The process is followed by a carbonization and a chemical activation in order to enhance the porosity, producing large number of fine pores. The tests of CO_2 capture showed that the optimized material achieved very high CO_2 capacity, among the highest reported for carbon-based sorbents.

In chapter 4, Dr Hassouna presents her research perspectives. Her competences in the domain of polymers and the design of high performance materials open the realm of possibility in many different fields. She explains that she recently launched a project in order to develop new electrodes, synthetized from renewable resources, for supercapacitor devices. Another domain concerns medical applications. The first project involves the design of biodegradable porous nanocomposite materials and the second project proposes to integrate active pharmaceutical ingredients (API) into polymer matrices in order to form amorphous solid dispersions (ASD). The aim of this second project is to improve the bioavailability of poorly soluble API. This study is conducted in collaboration with a laboratory of applied thermodynamics and represents an important issue.

Remark on the habilitation thesis

If compatible with the procedure of the University of Chemistry and Technology of Prague for habilitation thesis, a Curriculum Vitae would have been helpful to follow the scientific career of Dr Hassouna as well as some explanations on the context of each study (collaborations with other groups, fundings, supervised students…).

Conclusion

Dr Hassouna's research topics are varied, with a common focus on the design and/or optimization of polymeric materials for different applications. The diversity of the covered subjects enabled her to build herself a broad spectrum of knowledge in this field while remaining consistent with the environmental objectives announced. Dr Hassouna is undoubtedly a recognized specialist of PLA and its derivatives. Each study is well documented, detailed characterizations are systematically carried out and the results are published in good reviews. The work is solid and of a very good scientific level. The research activity carried out is original, the recent subjects developed are at the heart of current concerns and the proposed perspectives are of high interest.

I give a very favourable opinion for Dr Hassouna's habilitation thesis.

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