Synthesis of Molecularly Imprinted Polymers – Catalytic Properties of Obtained Materials

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PhD Thesis; 183 pp.

The PhD thesis submitted by Joanna Czulak concerns original research in the area of synthetic materials chemistry. It describes the design, synthesis, characterisation and evaluation of new catalysts produced *via* templating approaches. The templating is performed using a variety of methods of molecular imprinting. It is a broad-based, non-trivial body of work which includes organic chemistry, polymer synthesis and the characterisation and the evaluation of the catalytic properties of the materials produced. Whilst there are clear links/overlaps to other works reported in the open literature, the thesis represents a logical and interesting extension of the prior art.

The area of study is topical and entirely appropriate for a PhD study. The findings will be of interest to other scientists working in the area of synthetic materials chemistry, especially to scientists with a special interest in molecular imprinting and catalysis, but it will also have wider appeal.

The candidate sets her work in context in the **Abstract** (pp. 7-9) before launching into an introduction to the field (**Chapter 5**) and the aims of the research works (**Chapter 6**). Chapter 5 includes most of the keys areas that I would expect to be covered, draws particular attention to areas of the literature of particular relevance to the thesis work (*e.g.*, molecular imprinting and bioimprinting), and cites up-to-date, relevant references. Unfortunately, Chapter 5 is significantly under-referenced. Furthermore, there is an over-reliance on the citation of review articles; many of the leading, original citations are not referenced and this is a big oversight (examples include the imprinting works of Wulff, Mosbach and Shea, and the biocatalysis work of Klibanov). The historical perspective is also poorly delineated.

Chapter 7 and Chapter 8 describe the Materials and Methods, respectively. A range of interesting materials has been produced. In the majority of cases the chemical complexity is high, which makes structural characterisation difficult; nevertheless, reasonable efforts have been made in this regard, and the synthetic methods are mostly sufficiently detailed to enable these material preparations to be replicated in a different laboratory. There are some errors, *e.g.*, Table 7, p.55, no initiator, and it would have been preferable to have included the purity of the organic chemicals listed on pp. 48-53.

Chapter 9 is the Results & Discussion chapter, in which the results arising from the three distinct work packages (biomimimetics of metalloenzymes; HRP bioimprinting; HRP-imprinted polymer nanoparticles) are presented and discussed. I found this chapter to be particularly difficult to digest as a result of the style in which it was written; very often it was necessary to re-read passages and/or turn to Chapter 7 for cross-referencing purposes in order to understand and appreciate the commentary, and even then I was left with unanswered questions. This is unfortunate, because whilst the work is challenging and interesting in equal measure, and it most certainly advances the field of study, much more could have been made of the data. There were too many speculative comments not backed up with hard data, and the level of discussion and critical appraisal of the data was limited at times. In my opinion the thesis is probably publishable in part (although further data may be required).

The thesis closes with a summary of the General Observations and Future Work (**Chapter 10**) and a list of Final Conclusions (**Chapter 11**). Some suggestions for future work are presented here as well.

In terms of the overall quality of the presentation, the thesis was reasonably attractively produced, but not the easiest to read because of a large number of typographical and linguistic errors, some of which I have marked-up in my copy of the thesis. Fortunately, there were relatively fewer scientific errors, although basic errors with thermodynamics (p.14), chemical bonding theory and the consistent use of abbreviations stick in the mind. The tables and figures were presented reasonably clearly, and these aided the understanding of the material presented: indeed. further figures illustrate/emphasise/explain the new ideas/concepts/data would have been welcomed. The citations (Chapter 12) are not presented in a standard, consistent format.

Overall, then, whilst the thesis as written does have some deficiencies, I am confident that the work described within the thesis is original, of PhD standard and is publishable in part. On this basis, I am happy to recommend that the thesis proceed to further examination through *viva voce*.

Professor Peter Cormack University of Strathclyde 22nd February 2018

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