Alison McLellan

Corrections that have not been addressed in final document.

Comments from Dr Daniel Lee:

75. Surface species? What's the surface area?

The samples used in this chapter were prepared by a collaborator and they were not able to provide this information.

76. Why can T1 be measured and T2 not?

Long experiment times to obtain the variable temperature data and limited access to samples prevented a full suite of experiments being completed.

101. Why fig. 5.6b and fig. 5.7a so different? Reproducibility?

This has been actioned – I had noted the wrong composition of radical solution in figure 5.6b – this sample had a lower glycerol content.

104. Ion effects from Napa with the radical?

I believe that investigation of this falls outwith the scope of this study. The results presented are comparing the effect of changing the solution that is used to impregnate the sample. As the polymer and radical are constants, it is unlikely that any interaction between the polymer and radical will have a significant impact on these results.

104. Would be useful to show 13C NMR of pure powder.

There was very limited access to equipment capable of measuring good 'standard' ^{13}C spectra during my PhD so it was not possible to measure for all polymers used, I didn't want to include just some that I do have and not others.

107. How does Tdnp vary between samples? Where does depolarisation come into it?

T_{DNP} does not vary significantly, there is no significant correlation between T_{DNP} and SNR of MW off spectra. I could not measure T_{1e} for these samples so could not comment more on any depolarization.

108. Fig. 5.11 high intensity in a, low in b, vice versa

I don't quite understand this comment, in figure 5.11 the intensities have been normalized so that a comparison of trends can be seen rather than comparison of signal intensity.

129. temperature of scCO2 impregnation? Pressure...

I do not know which point of page 129 this is referring to, the impregnation temperatures and pressures are noted for all samples.

136. Add a solvent after scCO2 to see if that improves things?

This was discussed in the Viva, I did attempt this for a sample that I did not include in the thesis as the results were not of a high standard and I did not have sufficient time to test this further.

138. Significant loss of radical is opposite to what is stated above from EPR measurements

I don't think this is the case, the EPR results show that there is *some* radical present but I was unable to do any spin counting EPR experiments so the EPR results have no indication to the amount of radical in the samples.

155. How can intramolecular e-e interactions be removed as inter-mol. interaction dominate? This would look the same.

I do not fully understand this comment. The EPR calculations were done by Prof. Eric McInnes at the EPSRC National Service for Electron Paramagnetic Resonance Spectroscopy at The University of Manchester so I do not have full insight to the specific steps taken to remove the intra-molecular e-e interactions.

Comments from Dr Alexey Potapov:

55. The optoelectronic properties and efficiency of $MAPbI_3$ are greatly influenced by ion diffusion and migration. – "how? Why?"

A detailed explanation of the structure-property relationship of the materials is beyond the scope of this work and I don't believe necessary for the results presented. A more thorough description can be found in the references provided.

70. provide a numerical estimate for the diffusion rate (it would be a floor or ceiling number) based on your data.

It is not possible to estimate a diffusion rate due to the limited temperature range that was able to be investigated.

84. The DNP on polymers has been used quite long ago (late 80's - yearly 90's). Consider works by Schaefer, Yannoni and Wind.

Early work on DNP of polymers was previously reviewed as part of a more thorough literature review at the beginning of the project however I deemed that an overview of earlier work was not impactful here where the impact of the *knowledge gained* from DNP of polymers is the focus, rather than the ability to perform such experiments.

93. the size of the particles?

The swelling behaviour of the samples and swelling conditions used means it is unlikely that particle size would have a significant impact on results. Samples were left to equilibrate to ensure they were fully swollen regardless of particle size.

136. However, Le et al. show enhancments of 5-8

The work from Le et al. was published towards the end of this project so was not considered when designing the project. The results from their work are addressed from page 138.