Chapter 1

Introduction

Some general, overall aims and objectives of this study, as well as some introductory topics are described. More specific aims and introductions are stated within each draft article, which forms the chapters of this thesis.

1.1. The evolution of this project

The first and foremost aim of this project was to use the cheaper carbonate layered double hydroxide (LDH-CO_3) as a starting reagent in producing fatty acid intercalated LDHs, in stead of the more expensive and cumbersome to prepare chloride LDH. Ion exchange between the highly charged CO_3^{2-} and the sodium salt of a carboxylic (fatty) acid is not a viable process [1,2]. The fatty acid was used as starting reagent in stead of its salt. The pK_a values of carbonic acid are 6,35 and 10,33 at 25 °C respectively for the two protonation steps [3,4], which are higher than that of the fatty acids (4,8 at 25 °C [5]). The carbonate anion should be protonated by the fatty acid because an acid will donate a proton to the conjugate base of any acid with a higher pK_a [6]. CO_2 gas will subsequently be released. Hibino *et al*. [7] found that in LDHs with Mg/Al ratio of 2, no leaching of Mg^{2+} cations took place in acideous aqueous paramolybdate solutions, whereas substantial leaching of Mg^{2+} took place for LDHs with Mg:Al ratios of 3 and 4. Therefore one does not expect Mg^{2+} ions to leach during the intercalation reaction involving stearic acid and the LDH $Mg_4Al_2(OH)_{12}CO_3{\cdot}3H_2O$.

In the current study the intercalation reaction between stearic acid and the LDH was hampered by lump formation. Surfactants (such as SDS) were consequently added to adsorb onto the LDH surface and to emulsify the stearic acid. The performance of polymeric surfactants or polyelectrolytes such as poly(vinyl sulfonate) was tested and led to the discovery of a new route to intercalate polymers into the LDHs. Finally, the biodegradable film forming polyelectrolyte sodium alginate was used in the role of surfactant for the intercalation reaction. This led to the development of films with desirable physical and water vapour barrier properties.

1.2. The aims of this research

- To develop new techniques for intercalating organic anions (especially fatty acid anions, such as stearate) into LDH-CO_3 and to compare the new techniques to existing techniques in literature. The new technique should not use calcination steps or N_2 atmosphere.
- To develop new techniques for intercalating anionic polymers into LDH-CO_3 in order to produce nanocomposites.
- To use the modified LDH as a filler in thermoplastic starch composites in order to reduce the water vapour permeability of starch.

Chapter 2 describes the intercalation method we used in comparison to existing literature methods. Chapters 3 and 4 describe some applications of these stearate intercalated LDHs, namely as a starting reagent to easily intercalate anionic polymers and as a filler in starch-based composites.

References

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