

A Stepwise Route to the Synthesis of Mg-Al LDHs and the Intercalation Assembly of Organo-LDHs

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Abstract: Mg-Al layered double hydroxides (LDHs) was synthesized by stepwise route, and benzoic-LDHs, o-aminobenzoic-LDHs and o-hydroxybenzoic-LDHs intercalation assembly were obtained by ion exchange method. In the stepwise route, we controlled the state of Al composition as Al hydroxyl aqueous complexes in the aqueous solution first, then dropwise Mg^{2+} and OH^- and keep the procedure at constant pH value simultaneously at normal room temperature. The textural and structure properties of the resulting Mg-Al LDHs and organo-LDHs were characterized by XRD. The results showed that LDHs obtained were pure and highly crystalline. The intercalation assembly of benzoic-LDHs, o-aminobenzoic-LDHs and o-hydroxybenzoic-LDHs obtained by ion exchange kept the layer structure.

Key words: Mg-Al-LDHs; Al hydroxyl aqueous complexes; organo-LDHs; intercalation assembly

CLC number: O614 **Document code:** A **Article ID:** 0529-6579 (2007) S1-0285-02

Layered double hydroxide (LDHs) is a typical material with special two-dimensional layer structure^[1] and is a good matrix to design and obtain layered heterostructured hybrid materials^[2]. There are many reports about the synthesis of LDHs^[3] and their intercalation assembly^[4]. In this work, we established a stepwise route to synthesize Mg-Al LDHs. We controlled the state of Al composition as Al hydroxyl aqueous complexes in the aqueous solution first, then dropwise Mg^{2+} and OH^- and keep the procedure at constant pH value simultaneously at normal room temperature. Mg-Al LDHs was successfully synthesized by this route. Intercalation assemblies of organo-LDHs were obtained by ion exchange method studied.

1 The stepwise route to synthesis of Mg-Al LDHs and the intercalation assembly

First, the pHs of $Al(NO_3)_3$ aqueous solution was adjusted at different values by NaOH. Each resulting slurry or liquid with different pHs were dropwise with $Mg(NO_3)_2$. The product aged, then separated by repeated cycles of centrifugation and washing with deionized water three times and dried. Intercalation as-

sembly of organo-LDHs was obtained by the direct ion exchange. The procedure was held at constant temperature for several hours with stirring and refluxing, after the repeated cycles of centrifuging and washing three times with deionized water, and dried, the organo-LDHs samples were obtained.

2 Results and discussion

All the cases, excepted the pH value in the range of 7.0 ~ 8.0 (sample 1), LDHs phases were obtained, as observed from the XRD patterns showed in Figure 1. The purity and the crystallinity of the samples were very different. The morphology of Al composition in the synthesis system was mixture of $Al(OH)_3$ colloid and $Al(OH)_3$ slurry at pH 7.0 ~ 8.0. No Al ion coprecipitate with Mg^{2+} ion and OH^- . So there was no LDHs formation. As the pH value of the system was in the range of 8.5 ~ 12.5 (samples 2, 3, 4, 5, 6), the morphology of Al composition was mixture of Al hydroxyl aqueous complexes and $Al(OH)_3$ slurry. Al hydroxyl aqueous complexes ion coprecipitate with Mg^{2+} ion and OH^- . With the pH values increasing, the amount of Al hy-

①收稿日期: 2007-01-10

基金项目: 中国纤维材料改性国家重点实验室开放课题资助项目 (5G10620212); 郑州轻工业学院科研基金资助项目

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droxyl aqueous complexes increased. The yield of LDHs was increased and the MgO was decreased. When the pH value of the system was higher than 11.5, the product was pure and well-crystallized LDHs.

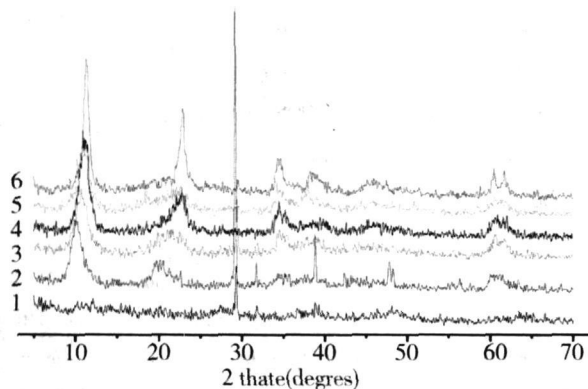


Fig 1 The patterns of XRD of samples of LDHs

The XRD patterns of intercalation assembly of organo-LDHs shows that the different organo-LDHs were synthesized by the direct ion exchange method. The three intense peaks in XRD patterns of organic LDHs at low diffraction angle shifted to the lower angles and the diffraction peak of (110) crystal plane is at the same diffraction angle. The interlayer space of LDHs was expanded by the intercalations of organic molecules and the brucite-like sheet had not any change.

3 Conclusions

The MgAl-LDHs was synthesized through the stepwise control route successfully. The pH value of the

synthesis was the key factor in the synthesis procedure. In the stepwise control route, as the pH value was greater than 8.5, Al hydroxyl aqueous complexes co-precipitated with Mg^{2+} to form LDHs in an alkaline solution. As the pH values of synthesis system in the range of 8.5 ~ 11.0, the products were mixtures of LDHs and MgO. And when the pH value of the synthesis system was higher than 11.5, the product was pure and well-crystallized LDHs.

Benzoic-LDHs, o-aminobenzoic-LDHs and o-hydroxybenzoic-LDHs intercalation assembly of organo-LDHs samples were obtained by the direct ion exchange.

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逐步法制备 MgAl-LDH与有机-LDH的插层组装体

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摘要: 通过控制铝组分在溶液中的形态, 采用逐步控制的方法制备了 MgAl-LDHs。经过离子交换, 制备了 benzoic-LDHs、o-aminobenzoic-LDHs 和 o-hydroxybenzoic-LDHs。在控制过程中, 铝的羟基配合物在室温下与 Mg^{2+} 和 OH^- 共沉淀生成 LDHs。XRD 图谱显示生成的 LDHs 晶相单一、结晶良好, 有机-LDHs 保持了层状结构。

关键词: MgAl-LDHs; 铝羟基配合物; organo-LDHs; 插层组装体

中图分类号: O614