



Intercalation, characterization and release behavior of imidacloprid into layered hydroxide salts by coupling of (3-glycidyoxypropyl) trimethoxysilane

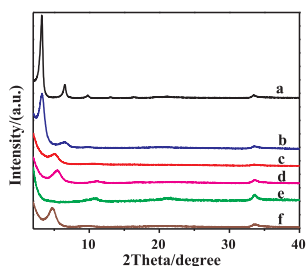


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GRAPHICAL ABSTRACT



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ABSTRACT

The poorly water-soluble pesticide imidacloprid (IMP) was intercalated into NiZn-layered hydroxide salts (NZL) by the aid of (3-glycidyoxypropyl) trimethoxysilane (GPS). The resulting NZL-GPS-IMP composites were characterized by X-ray diffraction (XRD), fourier transform infrared spectroscopy (FT-IR), carbon solid state nuclear magnetic resonance (¹³C NMR), thermo gravimetric analysis and differential thermal analysis (TGA/DTA). The interlayer distance of NZL-GPS-IMP was 1.62 nm, which is different from those of NZL-SDS, NZL-GPS and NZL-IMP. XRD and ¹³C-NMR results showed that SDS anions leave the galleries, and adsorb on the surface. The incorporation mechanism of NZL-GPS-IMP was reasonably put forward. TGA/DTA data showed that the combustion temperatures of IMP and GPS in the gallery were enhanced by 40 and 170 °C, respectively. These indicated that GPS-IMP was intercalated into the gallery. The IMP release behaviors of NZL-GPS-IMP in the water and buffer solutions (pH 5.0 and 6.8) were studied and compared. The IMP release rate and equilibrium release amount were dependent on release environments. Moreover, the release behavior of NZL-GPS-IMP was well fitted with pseudo second-order and parabolic diffusion models. This work would be helpful to develop a potential controlled-release formulation.

1. Introduction

Layered hydroxide salts (LHS) is a family of layered inorganic materials with positively charged layers and exchangeable interlayer anions [1,2], which have been widely concerned in the recent years. The

ion-exchange property of LHS is similar to layered double hydroxides (LDH). Layered zinc hydroxide nitrate (ZHN) salt has been usually investigated and applied in the fields of drug delivery [3–5]. Yang, et al. [3] have developed drug delivery system of indole-3-acetic acid-ZHN, and the release rate of the composite was slower than that of LDH.

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Moreover, pesticides of 4-(2,4-dichlorophenoxy)butyrate [4] and 2,4,5-trichlorophenoxyacetic acid [5], and drugs (ibuprofen, diclofenac [5], mefenamic acid and 4-biphenylacetic acid [6]) have been successfully intercalated into ZHN. The results showed that the composites may be applicable in controlled drug delivery systems. LHS may contain two types of cations in the octahedral sites of the layer, such as NiZn-layered hydroxide salts (NZL), which have been investigated as adsorbent [7], catalyst [8], and so on. In our previous works [9,10], it is interesting to find that NZL has much higher thermal stability than ZHN, and can be well synthesized in the pH range of 6.5–9.0.

It is well-known that organic compounds can be intercalated into LDH mainly through the electrostatic interaction or hydrophobic interaction. In addition, the silane coupling agent (3-aminopropyl) triethoxysilane has been intercalated into LDH by the covalent bonding [11]. Furthermore, (3-glycidyoxypropyl) trimethoxysilane (GPS) attached triazabicyclodecene has been also intercalated into LDH through the reaction of sodium dodecyl sulfate (SDS) molecules of SDS-LDH hybrid with cetyl trimethyl ammonium bromide (CTAB) molecules [12]. Whether can the pesticide be intercalated into LHS by the aid of the silane coupling agent or not? As far as we know, the similar research has not been reported until now. The hydrophobic pesticide imidacloprid (IMP) is an important neonicotinoid insecticide, which is used widely on rice, wheat, cotton, vegetables, fruiter tree, etc. The intercalation of the hydrophobic pesticide/drug is usually dependent on anionic surfactants [13–15] or the modified β -cyclodextrin [16–18]. NZL is selected as intercalation matrix in this study. The aim is to investigate the intercalation of IMP into NZL hybrid by means of GPS. First, NZL-SDS is prepared by the coprecipitation method; second, the solutions of IMP and GPS, CTAB are simultaneously added into NZL-SDS to obtain NZL-GPS-IMP. The structure properties of hybrid materials are determined by XRD, FT-IR, ^{13}C -NMR and TGA/DTA. The release behaviors of IMP from NZL-GPS-IMP are investigated and analyzed in different mediums.

2. Experimental

2.1. Materials

Sodium dodecyl sulfate (SDS) was purchased from Sinopharm Chemical Reagent Co., cetyl trimethyl ammonium bromide (CTAB) from Shanghai Mountain Pu Chemical Co., Ltd., China. (3-glycidyoxypropyl) trimethoxysilane (GPS) was purchased from Hubei Wuhan University Silicone New Material Co., Ltd., China. N, N-dimethylformamide (DMF) was purchased from Tianjin Benchmark Chemical Reagent Co., Ltd., China. Trichloromethane and acetone were obtained from Tianjin Chemical Reagent Third Factory, China. Imidacloprid (IMP) was bought from Jiangshu Changlong Chemical Co., Ltd., China. The structures of GPS and IMP are shown in Fig. 1. NaOH (analytical grade) was bought from North Tianjin Pharmacy Chemical Reagent, China, $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (analytical grade) are obtained from Tianjin Kemiou Chemical Co., Ltd., China. Decarbonated deionized water is used in the experiments.

2.2. Synthesis

SDS intercalated into NZL galleries (NZL-SDS) was prepared by coprecipitation method. Under a nitrogen atmosphere, 30 mL mixed solution ($0.5 \text{ mol L}^{-1} \text{ Zn}(\text{NO}_3)_2$ and $0.5 \text{ mol L}^{-1} \text{ Ni}(\text{NO}_3)_2$) and 20 mL

NaOH solution (0.5 mol L^{-1}) were simultaneously added dropwise into 20 mL of SDS solution (0.23 mol L^{-1}) under vigorous stirring at 30°C . The pH was fixed on about 6.5. Then, the mixture was hydrothermally treated at 100°C for 3 h in stainless-steel autoclave. The resulting sample was filtered and washed three times with decarbonated water, then dried at 85°C for 12 h and denoted as NZL-SDS.

Preparation of NZL-GPS-IMP was described as follows: firstly, 1.5 g IMP was solubilized into 5 mL DMF, followed by 15 mL GPS addition and stirred at 45°C for 24 h to obtain the solution 1; secondly, a certain weight CTAB was solubilized into trichloromethane to obtain the solution 2; finally solution 2 was added into solution 1, and followed by 1.0 g NZL-SDS addition and stirred at 45°C for 48 h. The mixture was filtered and washed three times with trichloromethane. The resulting solid sample was denoted as NZL-GPS-IMP (m), in which the $m = 0.2, 0.5$ or 0.7 , indicating the mass of the added CTAB. In addition, NZL-GPS and NZL-IMP samples were also prepared without IMP or GPS addition to make a comparison.

2.3. Characterization

Powder X-ray diffraction (XRD) were collected on D8 Focus powder diffractometer, using Ni filtered Cu $K\alpha$ radiation ($\lambda = 1.54056 \text{ \AA}$, 40 kV and 40 mA) between 2 and 80° at room temperature. FT-IR spectra ($4000\text{--}400 \text{ cm}^{-1}$) were recorded on a Tensor 27 spectrometer (Bruker, Germany) using fine round samples combined the KBr (spectroscopic grade) method with a sample/KBr mass ratio of 1: 100. Thermogravimetric analysis and differential thermal analysis (TGA/DTA) measurements were collected using a SDT Q600 synchronization thermoanalyzer (TA Instruments, New Castle, DE) in the range $20\text{--}800^\circ\text{C}$, using a heating rate of $10^\circ\text{C}/\text{min}$ under a flowing stream of air. UV–vis spectra were performed on TU1221 spectrophotometer. Nuclear magnetic resonance spectra of the hybrid materials were obtained on a Bruker Avance III 600 MHz Wide Bore spectrometer (Larmor frequency of 600.48 MHz and 151.01 MHz for ^1H and ^{13}C , respectively) equipped with 4 mm MAS probe, ZrO_2 rotor with MAS spin rate at 10 kHz. In order to increase the signal-to-noise ratio of the solid-state spectra, the CP/MAS technique was used. The ^{13}C CP/MAS spectra were recorded using variable amplitude ramped CP pulse of 2 ms. The recycling delay was 4 s.

2.4. Release studies

2.4.1. Determination of IMP loading

A given mass of NZL-GPS-IMP was placed in a 10 mL volumetric flask. 5 mL 1 mol L^{-1} HCl was added to dissolve NZL-GPS-IMP layers, and then ethanol was filled in the flask. The content of IMP in solution was monitored by the absorbance ($\lambda_{\text{max}} = 270 \text{ nm}$) on UV–vis spectroscopy. It was obtained according to the standard curve of IMP. All experiments were made in triplicate. The IMP loading was obtained by IMP content and used mass of NZL-GPS-IMP.

2.4.2. Determination of release behavior

The IMP release experiments were carried out at 30°C in pH 5.0 and 6.8 buffer solutions, respectively. 0.02 g NZL-GPS-IMP was dispersed in 250 mL buffer solution under magnetic stirring. 3 mL of supernatants were taken out at given time intervals and filtered, and then the same amount of fresh medium was added into solution. The IMP contents were determined by monitoring the absorbance ($\lambda_{\text{max}} = 270 \text{ nm}$), and

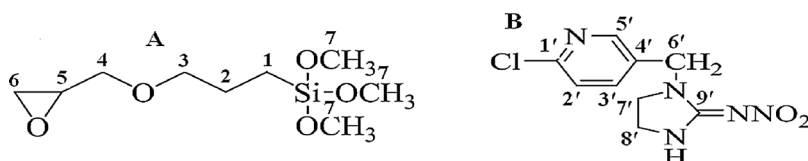


Fig. 1. Structures of GPS (A) and IMP (B).

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