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Mechanochemical synthesis of $FeSbO_4$-based materials from FeOOH and Sb_2O_5 powders

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Abstract

Iron antimony oxide ($FeSbO_4$) with specific surface area (SSA) over 50 m^2/g was synthesized mechanochemically by milling a mixture of iron oxy-hydroxide (FeOOH) and antimony pentoxide (Sb_2O_5) using a planetary ball mill at room temperature. The mechanochemical reaction proceeds with an increase in milling time and has been completed by 120 min. The prepared product powders are in the state of agglomerates consisted of fine particles of several dozen nanometers. This method has been extended to synthesis of $FeSbO_4$-based materials with different Fe/Sb atomic ratios ($1 \leq Fe/Sb \leq 4$). The SSA value of these prepared samples is in the range of 50 to 65 m^2/g.
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Keywords: Mechanochemical reaction; Milling; $FeSbO_4$; FeOOH; Sb_2O_5

1. Introduction

Iron antimony oxide ($FeSbO_4$) has been studied for utilization as (amm)oxidation catalysts for hydrocarbons [1–5] or carbon monoxide [6]. Especially, ammoxidation of propane to acrylonitrile is an interesting function of this material. Acrylonitrile has been generally produced by ammoxidation of propene, i.e. the Standard Oil of Ohio Company (SOHIO) process. In this process, the devices for dehydrogenation of propane are necessary to prepare propene. The use of the $FeSbO_4$ catalyst induces direct ammoxidation reaction of propane to acrylonitrile, and this leads to advantage for allowing no use of dehydrogenation device [4]. Another function of $FeSbO_4$-based materials lies in gas sensing. It is well known that SnO_2 is a high sensitive sensor material for liquid petroleum gas (LPG) [7–9]. However, selectivity of SnO_2 is not high [10,11], therefore, chemical modification is requested for elevating the selectivity [12–14]. Recently, Zhang et al. has reported the sensitive response to LPG by $FeSbO_4$-based materials with high sensitivity and selectivity compared with SnO_2-based materials [15–18]. $FeSbO_4$-based materials may become promising alternatives to SnO_2-based materials.

$FeSbO_4$ has been synthesized by mainly two routes: One is common ceramic powder elaboration at high temperature, i.e. solid state reactions between α-Fe_2O_3 and Sb_2O_3 at high temperature in the range of 1226 to 1246 K [19,20]. The product particle sizes prepared by this method are generally large so that they are low specific surface area (SSA). This may be a drawback in terms of a catalyst and a gas sensor. The other route is a wet process such as a co-precipitation method [4,6,16]. This method can provide fine and homogeneous product particles, however, the process is complex involving pH adjustment of solution, filtering and drying. In addition, the precursor prepared from the complex process needs to be calcined at high temperature above 808 K to form $FeSbO_4$ [16]. The SSA value of this product depends on the calcination temperature. For example, the SSA value is about 70 m^2/g at 823 K, while the calcination above 873 K induces rapid decrease in SSA value to less than 5 m^2/g [16]. There has been growing demand to develop a simple method for synthesizing a fine $FeSbO_4$.

Another route for preparing $FeSbO_4$ may be mechanochemical method. This method has a potential to form fine complex oxides in a single step milling operation at room temperature [21,22]. Recently, Berry and Ren has reported that $FeSbO_4$ can

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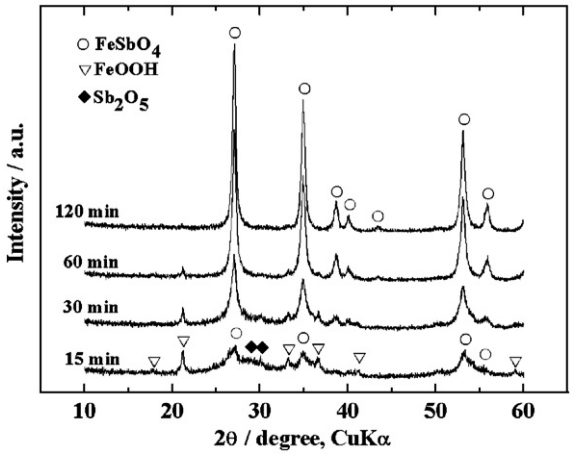

Fig. 1. XRD patterns of the FeOOH and Sb_2O_5 mixture milled for different periods of time.

be mechanochemically synthesized by milling a mixture of α-Fe_2O_3 and Sb_2O_3 [23]. In the method, Sb_2O_3 needs to be oxidized to Sb_2O_5 through the solid–gas reactions during the formation of $FeSbO_4$. Sb_2O_3 is more thermodynamically stable than Sb_2O_5, therefore, more energy for oxidization of Sb_2O_3 is required. In addition, α-Fe_2O_3 has corundum structure, which is known to be stable against milling operation [24–26].

We have attempted to synthesize $FeSbO_4$ by milling of FeOOH and Sb_2O_5 mixture at room temperature. One of the main purposes of this paper is to provide information on the physical properties of the product samples. In addition, according to the literatures, Fe/Sb atomic ratio in $FeSbO_4$-based materials is an important factor that influences their catalytic and sensing properties [5,17]. We have presented the fundamental data on the preparation of $FeSbO_4$-based materials with different Fe/Sb atomic ratios ($1 \le Fe/Sb \le 4$) by the milling in this paper.

2. Experimental

2.1. Samples and milling

Two kinds of sample were used for the synthesis of $FeSbO_4$-based materials: One is iron oxy-hydroxide (FeOOH) supplied from Kojundo Chemical Lab. Co., Ltd Japan, and the other is antimony pentoxide (Sb_2O_5) from Sigma-Aldrich USA. Both the raw materials, FeOOH and Sb_2O_5 were mixed with different Fe/Sb atomic ratios in the range of 1 to 4. Two grams of the mixture were put into a zirconia pot of 45 cm^3 inner volume with 7 zirconia balls of 15.7 mm diameter. A planetary ball mill (Fritsch Pulverisette-7, German) was used to mill the starting mixtures. The milling was operated in air at 700 rpm.

2.2. Characterization

The milled products were characterized by X-ray diffraction (XRD, RINT-2200, Rigaku, Japan) using CuKα radiation and Fourier transform infrared spectrometer (FT-IR, FTS3000, Digilab, USA) based on the standard KBr method to identify the phases formed after the milling. Morphology of the milled sample was observed by a scanning electron microscope (SEM, S4100-L, Hitachi, Japan). Specific surface area (SSA) of the samples was measured by using a nitrogen gas adsorption instrument (ASAP2010, Micromeritics, USA) based on the BET method. Before the SSA measurements, the samples were heated at 110 °C in vacuum for 5 h.

3. Results and discussion

3.1. Synthesis of $FeSbO_4$

Fig. 1 shows XRD patterns of the mixture of FeOOH and Sb_2O_5 with the Fe/Sb atomic ratio equal to 1.0, milled for different periods of time. In the pattern of the sample milled for 15 min, the phase identified as $FeSbO_4$ (JCPDS card No. 46-1387) is detected. As the milling time increases, the peak intensity of $FeSbO_4$ increases, and the single phase of $FeSbO_4$ is obtained by 120-min-milling, on the contrary, the peak intensity of FeOOH and Sb_2O_5 decrease. This indicates that $FeSbO_4$ can be formed by mechanochemical treatment. The reaction can be described as follows:

$$2FeOOH + Sb_2O_5 \rightarrow 2FeSbO_4 + H_2O \quad (1)$$

Fig. 2 shows IR spectra of the samples milled for different periods of time. According to the literatures, the bands at 901 and 804 cm^{-1} are ascribed to in-plane and out-of-plane Fe–O–H bending mode [27,28], respectively, and the band at 456 cm^{-1} corresponds to Fe–OH symmetric stretching vibration in FeOOH [28]. The band at 596 cm^{-1} may be assigned to Fe–O stretching in FeOOH [29]. With an increase in milling time, the intensity of these bands decreases, on the contrary, new bands appear around 700 and 530 cm^{-1}. These bands are attributed to the formation of $FeSbO_4$. The broad absorption band at 538 cm^{-1} can be ascribed to the stretching vibration of Sb–O and Fe–O bonds in MO_6 (M = Sb, Fe) octahedra. Both bands at 686 and 726 cm^{-1} can be attributed to stretching vibration of Sb–O bonds, which are in the typical rutile structure [19]. Both

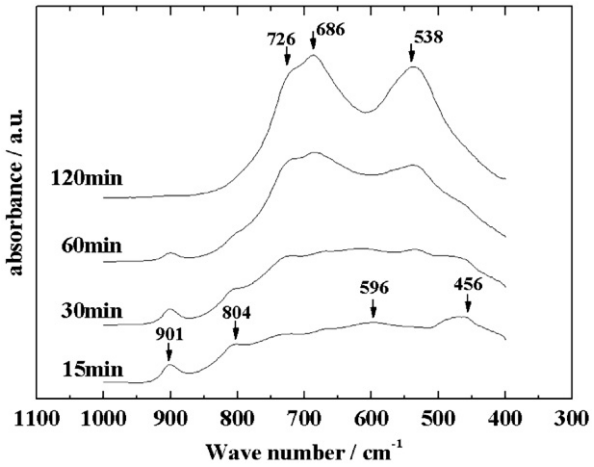

Fig. 2. Infrared spectra of the FeOOH and Sb_2O_5 mixture milled for different periods of time.

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