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Article

Catalytic decomposition of N₂O on cobalt substituted barium hexaferrites

Barkat UL-AIN ^{a,b}, Safeer AHMED ^b, HUANG Yanqiang ^{a,*}^a State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, Liaoning, China^b Department of Chemistry, Quaid-i-Azam University Islamabad, 45320, Islamabad, Pakistan

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ABSTRACT

Catalytic decomposition of N₂O was studied over Co substituted Mg-Y type hexaferrites (Ba₂Mg_{2-x}Co_xFe₁₂O₂₂, x = 0.4–2.0). The materials were synthesized by a sol-gel method using citric acid as the chelator followed by calcination at 1223 K for 5 h. X-ray diffraction, thermogravimetric analysis/derivative thermogravimetric, scanning electron microscopy, and N₂ adsorption techniques were used to confirm the formation of hexaferrite. The Ba₂Mg_{2-x}Co_xFe₁₂O₂₂ catalysts displayed good activity for decomposition of high concentration N₂O (30 vol%), and complete substitution of Mg for Co lowered the complete conversion temperature from 1123 K to 973 K. Increasing calcination temperature suppressed the catalytic activity by deteriorating surface area. Moreover, coupling microwave discharge significantly boosted the catalytic activity. Co substituted Mg-Y type hexaferrites are potentially active and thermally stable to be used as catalyst for high concentration N₂O decomposition.

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1. Introduction

Nitrous oxide (N₂O) is suggested as a green monopropellant for small satellite propulsion systems due to its low toxic nature and compatibility with the construction materials [1]. In satellite propulsion systems high concentration N₂O is used to produce as high propulsion power as possible. To be noted, decomposition of pure N₂O is a highly exothermic reaction having enthalpy of –82 kJ/mol and may lead to a temperature raise of more than 1600 K [2]. To find an active catalyst capable enough to decompose N₂O at low temperature is not a big deal. Many catalysts such as zeolite-based catalysts [3], perovskites [4], hydrocalcites [5,6], and supported noble metal catalysts [7] are extensively reported, but they cannot tolerate high temperature. In this situation the thermal stability is a much more

important factor evaluating practicability of any catalyst [8].

Hexaaluminates are a class of excellent high-temperature materials, which show stable phase composition up to 1900 K and high sintering resistance. The size of different large cations and introduction of transition metal ions could influence the structure of hexaaluminates, and then the catalytic activity in N₂O decomposition [9–11]. Hexaferrite also possesses layered structure, in which divalent and trivalent cations are distributed on octahedral and tetrahedral sites [12]. This arrangement endows the material with both high thermal stability and redox properties. Moreover, Y-type hexaferrite is a kind of microwave absorbing material and is widely used in microwave devices and magnetic recording media. Therefore, Y-type hexaferrite provides great potential as a N₂O decomposition catalyst. We reported previously that Al substituted Ba₂Mg₂Fe_{12-x}Al_xO₂₂ cat-

* Corresponding author. Tel: +86-411-84379416; Fax: +86-411-84685940; E-mail: yqhuang@dicp.ac.cn

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alysts exhibited desired N_2O decomposition activity. Especially under microwave heating mode [13], the complete decomposition of N_2O was obtained at 583 K without the use of expensive noble metals. However, their catalytic activity under conventional heating mode is still poor. In the present work, $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ was prepared and cobalt was introduced in structure to study its effect on catalytic activity. Effects of calcination temperature and microwave radiations on the catalytic activity were also studied for $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ ($x = 0.4\text{--}2.0$). The main aim of this study was to find out the appropriate composition of the material capable of decomposing N_2O at lower temperature under conventional heating mode while maintaining the novel decomposition capability under microwave irradiation mode.

2. Experimental

2.1. Catalyst preparation

A series of Co-substituted Y-type hexaferrites were prepared using a sol-gel route. For pure sample of $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$, 1.05 g of $\text{Ba}(\text{NO}_3)_2$, 1.28 g of $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, and 12.12 g of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ were dissolved individually in 100 ml of distilled water at 353 K and then added to 6.30 g of citric acid (SCRC 99.5%), which acts as the chelating agent. The pH of the solution was maintained at 8 by adding 2 mol/L $(\text{NH}_4)_2\text{CO}_3$ aqueous solution. A clear and transparent solution was obtained and was heated at 393 K for 3 h to get the wet gel. This gel was first pretreated at 723 K for 4 h to remove the unburnt impurities. The dry gel was then calcined at 1223 K for 5 h at the rate of 5 K/min to get the Y-type hexagonal phase of material. To study the effect of Co doping, several $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ catalysts were prepared with different Co content ($x = 0.4\text{--}2.0$) by the above mentioned procedure just by varying amounts of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ added.

2.2. Catalytic activity measurements

Activity measurements were carried out in a fixed-bed flow system. Before test, the catalyst was pretreated in Ar at 523 K for 2 h. Then, the reaction gas containing 30 vol% N_2O in Ar passed through 200 mg of a catalyst at a flow rate of 50 ml/min (STP), corresponding to a gas hourly space velocity (GHSV) of 30000 ml/(h·g). The bed temperature was increased from 723 K to 1073 K and held at each temperature for 30 min to reach a steady state. The effluent gas was analyzed by an on-line gas chromatograph (Agilent 6890N) equipped with a Chromosorb 103 column and a Molecular Sieve 13X column. N_2O conversion was calculated based on the difference between the inlet and outlet concentrations.

Microwave activity was measured by the microwave setup unit, details described elsewhere [13]. The reactor was filled with 200 mg of a catalyst for each run. An infrared pyrometer (Shanghai Institute of Technical physics, China) with a temperature range of 423–1273 K was used to measure the temperature of the catalyst bed. A thermocouple was used to recheck temperature measurement.

2.3. Catalyst characterization

BET surface areas of the catalysts were measured by N_2 adsorption at 77 K using a Micromeritics ASAP 2010 apparatus. Thermogravimetric analysis/derivative thermogravimetric (TGA/DTG) measurements were carried out on a Paulik-Paulik-Erdey type Q-1500 apparatus under the following conditions: 25 ml/min of O_2/He and a temperature ramp of 10 K/min up to 1273 K. The X-ray diffraction (XRD) patterns were recorded with a PANalytical X'Pert-Pro powder X-ray diffractometer, using $\text{Cu } K_\alpha$ monochromatized radiation, step-scan mode with a step size of $5^\circ/\text{min}$ and a counting time per data point of 10 s. The cell volume is dependent on the lattice parameters. Therefore lattice parameters were calculated by simultaneous equations development from XRD data. The obtained values were compared by the lattice parameters generated by computer software (JADE 9). The error bar in cell volume is obtained based on the difference between the obtained results. Crystallite dimensions were calculated using the Scherrer equation. Scanning electron microscopy (SEM) experiments were performed with a JSM 6360-LV electron microscope operating at 20–25 kV.

3. Results and discussion

3.1. Effect of cobalt loading on phase composition and catalytic performance

The TGA/DTG curves of the uncalcined gel precursor of $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ after heating at 393 K are depicted in Fig. 1. Several weight loss steps were observed upon heating the sample from ambient temperature up to 1273 K. A small weight loss took place at around 373 K, indicating the water evaporation. This is an endothermic process [14]. The second step weight loss occurred at 573 K, which can be due to the decomposition and rearrangement processes occurring within the sample with increasing temperature, including decomposition of iron nitrate to $\gamma\text{-Fe}_2\text{O}_3$, decomposition of barium nitrate to BaO, transformation of $\gamma\text{-Fe}_2\text{O}_3$ to $\alpha\text{-Fe}_2\text{O}_3$, and the reactions between iron oxides and BaO leading to mixed oxides, and decomposition of unreacted citric acid [15]. The third step weight loss at high temperature is due to the formation of oxides from

Fig. 1. TGA/DTG curves of the $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ sample after heating at 393 K.

Fig. 2. XRD patterns of the $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ catalysts with different cobalt contents ($x = 0.0$ – 2.0) calcined at 1223–1473 K.

hydroxides and mono-hexaferrites, and rearrangement to form pure hexaferrites. After 1173 K, there is no weight loss, indicating the complete formation of Y-type hexaferrites around this temperature.

XRD is employed for phase identification and to calculate structural parameters. Magnetoplumbite phase is prominent in all of the samples (Fig. 2). However, impurity phase of BaFe_2O_4 can also be observed depending on the amount of cobalt doping. All peaks of substituted and un-substituted samples match well with the standard pattern indicated by JCPDS 44-0206 and the main peaks are indexed. These XRD patterns also indicate that 1223 K is the appropriate temperature to get Y-type hexaferrites phase. These indexed peaks are further used to calculate the lattice parameters, crystallite size, and cell volume (Table 1). Introduction of cobalt does not change the phase of the material, indicating that cobalt can successfully replace magnesium. The ionic radius of Co^{2+} is 0.074 nm, which is comparable to Mg^{2+} (0.072 nm), so it does not affect the lattice parameters so remarkably as cell volume. The crystallite size of the samples ranges from 45 to 57 nm. To be noted, there was a slight increase of BET surface area with increasing the content of Co, as shown in Table 1.

Morphology of the prepared samples was observed by SEM. Figure 3 shows the SEM images of the pure and substituted samples. These micrographs show that all samples have crystalline nature. The homogeneous phase is evident with uniform particle distribution. For the pure sample most of the particles are rod like, but as the content of cobalt increases the particles become plate like and then finally attain the distorted hexagonal symmetry. No aggregation of samples is observed, which indicates the similar surface area.

Figure 4 gives the conventional heating profile of N_2O decomposition over the pure and substituted catalysts. It is clear that by increasing the cobalt content the catalyst activity significantly increased, and T_{100} (temperature at which 100% decomposition takes place) decreased from 1123 K to 973 K. Catalytic activity of ferrite is greatly dependent on the degree of cationic substitution and preparation conditions. There are

Table 1

Structural parameters of Co doped $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$.

Co content (x)	Calcination temperature (K)	a (nm)	c (nm)	V_{cell} (nm^3)	D (nm)	A_{BET} (m^2/g)
0.0	1223	0.592	4.368	1.326 ± 0.0004	54 ± 2	3 ± 0.5
0.4	1223	0.592	4.362	1.323 ± 0.0002	45 ± 3	4 ± 0.8
0.8	1223	0.589	4.369	1.312 ± 0.0005	57 ± 2	4 ± 0.6
1.2	1223	0.586	4.364	1.297 ± 0.0008	53 ± 4	4 ± 0.5
1.6	1223	0.583	4.358	1.282 ± 0.0006	56 ± 5	6 ± 0.4
2.0	1223	0.583	4.357	1.282 ± 0.0003	55 ± 3	8 ± 0.2
2.0	1273	0.588	4.362	1.306 ± 0.0002	53 ± 6	8 ± 0.3
2.0	1373	0.583	4.356	1.282 ± 0.0002	65 ± 2	4 ± 0.4
2.0	1473	0.583	4.358	1.282 ± 0.0004	67 ± 4	2 ± 0.5

only two symmetry types of interstitial sites present in Y-type hexaferrite available for metallic ions, namely six octahedral sites and two tetrahedral sites. Cation distribution on crystallographic octahedral and tetrahedral sites is very important in this regard. Tetrahedral sites are not exposed, so they are almost inactive crystallographic site. However, octahedral sites are more external, shared with adjacent spinel blocks, and lie on vertical threefold axis [16], thus contributing to catalytic activity. Hexaferrites based on 3d transition metals have been subject of applied research due to their catalytic properties [17]. Mg is non-transition element and it occupies the tetrahedral site in the structure. As far as Co is concerned, it belongs to 3d transition element, and it can exist in the oxidation states of +II and +III. Cobalt can occupy tetrahedral as well as octahedral

Fig. 3. SEM images of the $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ catalysts with different cobalt contents: (a) $x = 0.0$; (b) $x = 0.4$; (c) $x = 0.8$; (d) $x = 1.2$; (e) $x = 1.6$; (f) $x = 2.0$.

Fig. 4. N_2O conversion as a function of temperature over the $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ catalysts with different cobalt contents. (1) $x = 0.0$; (2) $x = 0.4$; (3) $x = 0.8$; (4) $x = 1.2$; (5) $x = 1.6$; (6) $x = 2.0$.

sites. Detailed literature survey reveals that many scientists relate the activity of cobalt based catalyst to the $\text{Co}^{2+}/\text{Co}^{3+}$ redox couple [18–20]. Co in its oxide forms has excellent catalytic property due to d^7-d^6 electronic configuration for Co^{2+} - Co^{3+} ions. This ion pair is reported to possess excellent catalytic activity for N_2O decomposition [21]. Abu-Zied et al. [22] postulated reaction mechanism to explain oscillations during N_2O decomposition on Co-ZSM-5 catalysts, and they also related the activity to redox properties of system $\text{Co}^{2+}/\text{Co}^{3+}$, where N_2O acts as a reductant or oxidant based on nature of active site. This redox cycle also encourages the formation of more oxygen vacancies resulting in better adsorption and decomposition.

In order to assess the stability of the catalysts, the N_2O conversion was monitored with time on stream. It is evident from Fig. 5 that the $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ catalyst exhibited stable catalytic activity for 24 h, and no deactivation was observed during this period. The desired catalytic activity and good stability of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ make it a potential candidate for high concentration N_2O decomposition.

3.2. Influence of calcination temperature on catalytic activity of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ for N_2O decomposition

Catalytic activity of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ calcined at different

Fig. 5. N_2O conversion at 873 K as a function of time-on-stream over the $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ catalyst.

Fig. 6. Activity of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ calcined at different temperatures. (1) 1273 K; (2) 1373 K; (3) 1473 K.

temperatures is shown in Fig. 6. The N_2O conversion decreased when the calcination temperature was raised. The sample calcined at 1473 K showed the worst catalytic activity. Figure 7 shows the XRD patterns of these three samples. The XRD patterns consisted of only hexaferrites phase. Increasing calcination temperature had no effect on the XRD patterns except that the intensity of the peaks got intensified, indicating the sintering of the catalyst samples. Figure 8 shows the SEM micrographs of the three samples. These micrographs show that more and more particles aggregated as treatment temperature was elevated. The uniform crystalline phase got distorted and there was a grain growth. The same trend is shown by BET surface area, and as the calcination temperature increases the surface area of the catalyst decreases (Table 1). The BET surface areas were 8, 4, and $2 \text{ m}^2/\text{g}$ for the samples calcined at 1273, 1373, and 1473 K, respectively. Surface area is a very important factor in determining the catalytic activity of the samples. These results indicate that catalyst sintering occurred when the calcination temperature was raised, leading to the decrease in the N_2O conversion (Fig. 6). SEM micrographs clearly show the aggregation of particles at high temperature. This aggregation leads to the decline in active sites for adsorption of N_2O molecules, which is responsible for suppressed catalytic activity.

Fig. 7. XRD patterns of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ calcined at 1273 K (1), 1373 K (2), and 1473 K (3).

Fig. 8. SEM images of $\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ calcined at 1273 K (a), 1373 K (b), and 1473 K (c).

3.3 Effect of microwave irradiation on catalytic activity of $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$

Remarkable results have been obtained by the use of microwave heating to stimulate catalytic reactions [23–25]. Hexaferrites are an excellent microwave absorbing material [26,27], and microwave discharge can enhance the catalytic activity to a significant extent. Figure 9 shows that coupling microwave discharge with prepared catalyst reduces the T_{100} from 1123 K to 583 K. This is a significant result because complete N_2O decomposition can be reached at such low temperature without using any costly noble metals. The mechanism of microwave catalysis is not so clear up to the date, but many propositions have been put forward. Hexaferrite is not only a good microwave absorber but also a material for nitrous oxide adsorption [28]. The high frequency radiations are effective for the activation of polar molecules, so it can be contemplated that N_2O adsorbed on hexaferrite could get activated by these electromagnetic radiations. This microwave discharge produces many active electrons and radicals having high energy and temperature. Therefore, although average temperature of the feed gas still remains low in the microwave discharge mode, N_2O decomposition takes place. Substituting magnesium with cobalt has not so much effect on the microwave activity with start over temperature and T_{100} being the same for all of the samples. There is even a slight decrease in catalytic activity by

the cobalt substitution. This is mainly due to the relatively low magnetic and dielectric losses and, in particular, the high resistivity [29]. For most ceramic magnetic materials used in microwave technology, the magnetic anisotropy field should be sufficiently small [30]. Substitution of cobalt results in an increase of in plane magnetic anisotropy [31] and slightly deteriorates the catalytic activity in microwave discharge mode.

4. Conclusions

Cobalt substituted $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ samples were explored for high concentration N_2O decomposition. Co can successfully replace magnesium without any effect on the crystal structure of Y-type hexaferrite. Substitution of Co enhances the catalytic activity by increasing the redox reaction between Co^{2+} and Co^{3+} . Increasing calcination temperature suppresses the catalytic activity by decreasing catalyst surface area and promoting aggregation. Coupling microwave discharge with prepared samples provides remarkable results by decomposing this gas at very low temperature of about 583 K. In a nutshell the Co substituted hexaferrite demonstrated to be a promising candidate for high concentration N_2O decomposition.

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Fig. 9. N_2O conversion over $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ with different Co contents in the presence of microwave discharge with power ranging from 10 W to 60 W. (1) $x = 0.0$; (2) $x = 0.4$; (3) $x = 0.8$; (4) $x = 1.2$; (5) $x = 1.6$; (6) $x = 2.0$.

Graphical Abstract

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Barkat UL-AIN, Safeer AHMED, HUANG Yanqiang *

Dalian Institute of Chemical Physics, Chinese Academy of Sciences, China; Quaid-i-Azam University Islamabad, Pakistan

Co substituted barium hexaferrites displayed good activity for N_2O decomposition. Complete substitution of Mg for Co lowered the complete conversion temperature from 1123 K to 973 K.

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Co取代的六铁酸盐在 N_2O 催化分解反应中的应用

Barkat UL-AIN^{a,b}, Safeer AHMED^b, 黄延强^{a,*}

^a中国科学院大连化学物理研究所催化基础国家重点实验室, 辽宁大连116023

^b伊斯兰堡奎德-i-阿扎姆大学化学系, 巴基斯坦伊斯兰堡45320

摘要: 采用溶胶-凝胶法制备了Co取代的 $\text{Ba}_2\text{Mg}_{2-x}\text{Co}_x\text{Fe}_{12}\text{O}_{22}$ ($x = 0.0 \sim 2.0$) 催化剂, 并考察了其在高浓度 N_2O (30 vol%) 分解反应中的催化活性. 结果表明, Co的取代显著提高了 $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ 催化剂活性, 当 $x = 2$ 时, N_2O 的完全分解温度从1123 K降至973 K; 提高催化剂焙烧温度会导致其表面积降低, 进而降低其催化活性; 但采用微波加热则能显著提高催化剂活性.

关键词: 一氧化二氮; 分解; 六铁酸盐; $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$; 钴; 微波

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*通讯联系人. 电话: (0411)84379416; 传真: (0411)84685940; 电子信箱: yqhuang@dicp.ac.cn

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