Preparation and characterization of R$_2$CoMnO$_6$ (R=La, Nd) via PVA sol-gel route

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1. Introduction

The double perovskite oxides, which was usually expressed in the form A$_2$BB'O$_6$ (where A represents lanthanide ion and B and B' represents transition metals ions) have attracted much attention in recent decades due to their magnetoresistance effect, magnetodielectric properties, magnetocaloric effect, multiferroic properties, and catalytic properties [1–5]. Among them, La$_2$CoMnO$_6$ has received special attention recently. La$_2$CoMnO$_6$ is a ferromagnetic insulator with a Curie temperature of about 225 K [6]. The substitution of the La ion by another rare earth element with a smaller ionic radius significantly changes their magnetic structure and properties [7]. As the ionic radius of the rare earth element decreases, the ferromagnetic Curie temperature decreases gradually, along with the structural distortion [8–11]. In addition, different synthesis methods resulted in complicated crystal structures [6,12]. As we can see, the conventional solid-state reaction method is commonly employed in the preparation of R$_2$CoMnO$_6$ (R = La, Nd) at high temperature [8,11,13,14]. In the meantime, the wet chemical synthesis of R$_2$CoMnO$_6$ was also reported. Various wet chemical methods, such as citric acid sol-gel method [7,12,15–19], ethylene glycol sol-gel method [4,20,21], polymeric precursor method [6,22], glycine-nitrate combustion method [23–26], urea-nitrate combustion method [27], nitrate decomposition method [9,10,28–30], etc. have been employed to prepare R$_2$CoMnO$_6$. For example, Dass and Goodenough [6] synthesized La$_2$CoMnO$_6$ using the polymeric precursor method, and the formation conditions of single-phase La$_2$CoMnO$_6$ was 600°C for 12 h. Liu et al. [21] reported the synthesis of phase pure La$_2$CoMnO$_6$ via ethylene glycol sol-gel method at 600°C. Silva et al. [22] synthesized monophasic La$_2$CoMnO$_6$ by polymeric precursor method at 900°C for 16 h. Joy et al. [23] have prepared LaMn$_{0.5}$Co$_{0.5}$O$_3$ using the glycine-nitrate method and single-phase LaMn$_{0.5}$Co$_{0.5}$O$_3$ was obtained at 500°C for 12 h. They [24] also reported the synthesis of NdMn$_{0.5}$Co$_{0.5}$O$_3$ using the glycine combustion method. Even calcined at 700°C for 12 h, a peak from hexagonal Nd$_2$O$_3$ can still be found in the XRD patterns, although the authors attributed the peak to superlattice reflection [24]. Sayed et al. [26] synthesized La$_2$CoMnO$_6$ by glycine combustion method. The obtained powders were amorphous and monophasic La$_2$CoMnO$_6$ can be formed at 650°C for 3 h [26]. La$_2$CoMnO$_6$ was also reported to be synthesized by the molten-salt method and single-phase La$_2$CoMnO$_6$ was obtained at 700°C for 6 hours [31,32]. Obviously, La$_2$CoMnO$_6$ can be prepared by various methods, but the preparation temperatures are relatively higher. On the other hand, the report on the wet chemical synthesis of Nd$_2$CoMnO$_6$ is very few. Low-temperature synthesis of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ can save energy. In addition, La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ powders synthesized at low temperatures have smaller particle sizes and may show properties different from those of the bulk, for example, certain ferromagnetic nanoparticles show superparamagnetism. Therefore, it is necessary to synthesize La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ at low temperature. The aim of this work is to synthesize La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ at low temperatures by a different method.

As we know, the PVA sol-gel method has been applied in the preparation of various metal oxide powders [33–35]. In this work, La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ were synthesized with the PVA route. Nanocrystalline La$_2$CoMnO$_6$ with average crystallite size ~20 nm was directly obtained during the charring procedure, while single-phase, nanosized Nd$_2$CoMnO$_6$ powders was synthesized at 600°C.
2. Experimental

La$_2$O$_3$, Nd$_2$O$_3$, Co(NO$_3$)$_{2\times}$·6H$_2$O, MnCO$_3$, and PVA (MW = 79000) were used as starting materials. The sol-gel preparation procedure of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ is as follows: First, 5 wt% PVA solution and nitrate solution were prepared, respectively. The 5 wt% PVA aqueous solution was made by dissolving PVA powder into deionized water at 80°C. On the other hand, La$_2$O$_3$ or Nd$_2$O$_3$ as well as MnCO$_3$ powders were dissolved in nitric acid, along with the addition of stoichiometric Co(NO$_3$)$_{2\times}$·6H$_2$O to form nitrate solution. Afterward, the above two solutions were mixed with stirring at 80°C until the transparent gel was formed. For 1 mol La$_2$CoMnO$_6$ or Nd$_2$CoMnO$_6$, a 6 mol PVA monomer was used [35]. The samples were charred in an oven at 250°C for 2 h and the obtained powders were referred to as precursor. Finally, the precursors were calcined at 500°C to 700°C for 2 h.

The thermal decomposition behavior of the precursor was studied by applying differential scanning calorimetry (DSC) and thermogravimetric (TG) analysis on STA 449 F3 Jupiter (Netzsch, Germany) with a heating rate of 10°C/min. The Fourier transform infrared spectra of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ precursor and powders were collected using Vertex 70 FT-IR spectrometer (Bruker Optik GmbH, Ettlingen, Germany). The phase purity of the obtained powders was characterized using X’Pert PRO X-ray diffractometer (PANalytical B.V., Almelo, Netherland) with Cu K$_\alpha$ radiation. The Raman spectra were collected at room temperature with 532 nm Nd:YAG laser excitation using a LabRAM HR800 single-stage spectrometer (Horiba JobinYvon Ltd., France). The morphology and size analysis of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ powders was carried out by field emission scanning electron microscope (FESEM) (GeminiSEM 300, Carl Zeiss, Germany). A physical property measurement system (PPMS) from Quantum Design (San Diego, CA) was used to study the magnetic properties of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ powders with magnetic fields up to 2 T.

3. Results and discussions

Figure 1 displays the simultaneous DSC and TG curves of La$_2$CoMnO$_6$ precursor. The DSC curve shows no obvious exothermic or endothermic peak, indicating that the reaction of forming La$_2$CoMnO$_6$ is complete. The later XRD patterns show that phase pure La$_2$CoMnO$_6$ has formed. A slight weight loss of 7.7% can be observed in the TG curve until the temperature reaches 1000°C, for the reason that the adsorbed moisture is gradually evaporated and residual organics is volatilized during the heating process.

The results of simultaneous DSC and TG for Nd$_2$CoMnO$_6$ precursor are presented in Figure 2. A small exothermic peak around 292°C in the DSC curve can be observed, which is associate with the oxidative decomposition of the majority of the organic material. On the other hand, the TG curve shows two-weight losses. The first weight loss of 5.4% below 100°C may associate with the vaporization of adsorbed moisture and the second weight loss of 4.6% between 100°C and 650°C can be attributed to the combustion of organics and the release of gases. After the temperature increased to more than 650°C, the sample weight gradually stabilized and remains almost unchanged.

Figure 3 displays the XRD patterns of La$_2$CoMnO$_6$ precursors and powders calcined at 500–700°C for 2 h. It can be seen that the single-phase La$_2$MnCoO$_6$ is formed directly in the precursor, and no peak from the unexpected intermediate phase is observed. All the diffraction peaks can be assigned to the monoclinic crystal structure [6], as indexed in the figure. The XRD patterns of La$_2$CoMnO$_6$ precursor calcined at temperatures ranging from 500 to 700°C. The XRD patterns of La$_2$CoMnO$_6$ precursor calcined at temperatures ranging from 500 to 700°C.
from 500°C to 700°C for 2 h showed no significant change, except a slight-increased intensity, indicating an increase in crystallinity. The crystallite size of La$_2$CoMnO$_6$ powder was estimated using the Scherrer formula: the crystallite size of La$_2$CoMnO$_6$ precursor and powders calcined at 500°C, 600°C, and 700°C for 2 h is about 20.8, 21.2, 22.3, and 23.5 nm, respectively. Table 1 summarizes the formation condition of single-phase La$_2$CoMnO$_6$ via various synthesis methods. We can see that the PVA sol-gel method in the present work has a lower temperature and shorter time for the synthesis of La$_2$CoMnO$_6$. To the best of our knowledge, this is the reported lowest synthesis temperature for La$_2$CoMnO$_6$.

The XRD patterns of Nd$_2$CoMnO$_6$ precursors and powders calcined at 500–700°C for 2 h are depicted in Figure 4. The XRD pattern of the Nd$_2$CoMnO$_6$ precursor shows that it is amorphous, which is different from that of La$_2$CoMnO$_6$. La$_2$CoMnO$_6$ phase formed directly in the precursor, indicating the formation temperature of single-phase Nd$_2$CoMnO$_6$ is higher than that of La$_2$CoMnO$_6$, which can only be attributed to the smaller radius of Nd than that of La. The Nd$_2$CoMnO$_6$ powder calcined at 500°

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Table 1. The preparation method and formation conditions of single-phase La$_2$CoMnO$_6$.

| Synthesis method | Formation condition | Reference |
|------------------|---------------------|-----------|
| Polymeric precursor | 600°C, 12 h | [6] |
| Polymeric precursor | 900°C, 16 h | [22] |
| Ethylene glycol | 600°C, not provided | [21] |
| Glycine | 500°C, 12 h | [23] |
| Glycine | 630°C, 2 h | [26] |
| Molten-salt | 700 °C, 6 h | [32] |
| PVA | 250°C, 2 h | This work |

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Figure 2. DSC-TG curves of Nd$_2$CoMnO$_6$ precursor via PVA sol-gel route.

Figure 3. XRD patterns of La$_2$CoMnO$_6$ precursors and powders calcined at various temperatures for 2 h via PVA sol-gel route.
C is still amorphous although some very weak peaks from Nd$_2$CoMnO$_6$ appeared. Calcining the precursor at 600°C for 2 h, monophasic Nd$_2$CoMnO$_6$ with monoclinic structure was formed. Moreover, when the calcination temperature reaches to 700°C, the XRD patterns still remain almost unchanged. The XRD patterns are consistent with those reported in Ref. 24 and 36. Therefore, it is convinced that phase pure Nd$_2$CoMnO$_6$ can be prepared at 600°C for 2 h by PVA sol-gel method. The crystallite size of Nd$_2$CoMnO$_6$ powder calcined at 600°C and 700°C for 2 h calculated from Scherrer formula is about 20.1 and 23.9 nm, respectively. There are very few reports on the wet chemical synthesis of Nd$_2$CoMnO$_6$. Joly et al. prepared NdMn$_0.5$Co$_0.5$O$_3$ using the glycine-nitrate combustion method. For the sample calcined at 700°C for 12 h, there may also be a small amount of hexagonal Nd$_2$O$_3$ impurity [24]. We can conclude that the PVA sol-gel method can synthesize single-phase Nd$_2$CoMnO$_6$ at a lower temperature than the glycine-nitrate combustion method.

Figure 4 presents the XRD patterns of Nd$_2$CoMnO$_6$ precursor and powders calcined at various temperatures for 2 h via PVA sol-gel route.

Figure 5. FT-IR spectra of La$_2$CoMnO$_6$ precursor and powders calcined at 500–700°C.
corresponds to nitrate ions \([35,36]\) and the formation of \(\text{La}_2\text{CoMnO}_6\) \([37]\), respectively. This is in accordance with the previous XRD result: \(\text{La}_2\text{CoMnO}_6\) phase has formed in the precursor (see Figure 3). After heating the \(\text{La}_2\text{CoMnO}_6\) precursor at 500–700°C, the intensities of absorption bands at 601 cm\(^{-1}\) increase due to increased crystallinity, and the band at 1384 cm\(^{-1}\) disappears because of the decomposition of residual nitrate.

The FT-IR spectra of \(\text{Nd}_2\text{CoMnO}_6\) precursor and calcined powders are presented in Figure 6. For the \(\text{Nd}_2\text{CoMnO}_6\) precursor, the absorption bands around 3436 cm\(^{-1}\) and 1631 cm\(^{-1}\) are related to \(\text{H}_2\text{O}\) molecules on the surface of the precursor; the absorption band around 1510 and 1385 cm\(^{-1}\) can be assigned to carbonate ions \([38,39]\). In the spectrum of \(\text{Nd}_2\text{CoMnO}_6\) calcined at 500°C for 2 h, no obvious change was observed except decreased intensities of carbonate and adsorbed water. Heating the precursor at 600°C for 2 h, two new strong absorption bands at 589 cm\(^{-1}\) and 450 cm\(^{-1}\) appears, suggesting the formation of \(\text{Nd}_2\text{CoMnO}_6\) \([37]\). Similarly, this is also in accordance with the XRD results in Figure 4. Compared with \(\text{La}_2\text{CoMnO}_6\), the substitution of \(\text{La}\) by \(\text{Nd}\) resulted in a shift of the corresponding absorption band to a lower wave-number. The FT-IR spectrum for the powder calcined at 700°C is almost the same as that of the powder calcined at 600°C.

Figure 7 displays the Raman spectra of \(\text{La}_2\text{CoMnO}_6\) precursor and powders calcined at 500°C and 700°C for 2 h. As can be seen from the figure, two Raman active modes around 515 cm\(^{-1}\) and 642 cm\(^{-1}\) are observed. The strongest Raman peak around 642 cm\(^{-1}\) can be attributed to the symmetric stretching of the basal oxygen ions of the octahedra \(\text{B} \approx \text{O} \approx \text{B} \approx \text{O}\) \([40]\). The second Raman peak around 515 cm\(^{-1}\) can be attributed to the antisymmetric stretching \((\text{A}_\text{g})\) associated with the Jahn–Teller distortion \([41–43]\).

These Raman spectra are consistent with that reported in Refs. 10, and 44–46. In addition, the obtained Raman spectra shows an increase in the intensity of the peaks as the calcination temperature grows higher, which implies that the crystallinity of \(\text{La}_2\text{CoMnO}_6\) increases.

The Raman spectra of \(\text{Nd}_2\text{CoMnO}_6\) precursors calcined at 500°C and 700°C are shown in Figure 8. The Raman spectra are similar to that of \(\text{La}_2\text{CoMnO}_6\) in Figure 7, but the two Raman peaks move to lower wavenumber, which is consistent with that reported in Refs. 10 and 36. The peaks around 627 and 475 cm\(^{-1}\) belong to symmetric stretching and antisymmetric stretching/bending modes of the \(\text{Co(Mn)O}_6\) octahedron \([40–42]\). Well-crystallized \(\text{Nd}_2\text{CoMnO}_6\) formed at 700°C, its Raman spectrum is strong. Very weak crystallization peak appears in the XRD pattern of \(\text{Nd}_2\text{CoMnO}_6\) precursors calcined at 500°C (Figure 4), its Raman spectrum is obviously weaker than that of \(\text{Nd}_2\text{CoMnO}_6\) precursors calcined at 700°C, but the two main Raman peaks have appeared.

The FESEM images of \(\text{La}_2\text{CoMnO}_6\) precursor, \(\text{La}_2\text{CoMnO}_6\) and \(\text{Nd}_2\text{CoMnO}_6\) powders calcined at 600°C for 2 h with different magnifications are presented in Figure 9. \(\text{La}_2\text{CoMnO}_6\) and \(\text{Nd}_2\text{CoMnO}_6\) powders show three-dimensional network structures. This is because, during the formation of the gel, the long-chain structure of PVA is interlaced to form a complex network structure in the space. The gel network is filled with a solvent. When the gel is dried and calcined to form \(\text{La}_2\text{CoMnO}_6\) or \(\text{Nd}_2\text{CoMnO}_6\) powders, the powders also present a spatial three-dimensional network structure. The enlarged images \(\text{(Figure 9 \(b,d,e)\)}\) revealed that the obtained powders are composed of smaller primary particles having a size on the order of nanometers.

The magnetization vs. applied magnetic field curves at room temperature and 100 K for \(\text{La}_2\text{CoMnO}_6\) precursor,
La$_2$CoMnO$_6$ powder calcined at 500°C, and Nd$_2$CoMnO$_6$ powders calcined at 600°C are shown in Figure 10 and 11, respectively. A linear relationship between magnetization vs. applied magnetic field at room temperature is observed for La$_2$CoMnO$_6$ precursor, La$_2$CoMnO$_6$, and Nd$_2$CoMnO$_6$ powders, indicating that the powders are paramagnetic at room temperature. However, La$_2$CoMnO$_6$ precursor, La$_2$CoMnO$_6$, and Nd$_2$CoMnO$_6$ powders show typical hysteresis loop at 100 K. This is consistent with the results reported in Refs. 7, 15, 27, and 36. Compared with La$_2$CoMnO$_6$ precursor, La$_2$CoMnO$_6$ powder calcined at 500°C shows larger remnant magnetization ($M_R$) and coercive field ($H_c$). The magnetization curves of Nd$_2$CoMnO$_6$ powders do not show any saturation at 100 K even at the highest magnetic field of 20 kOe. In addition to that, Nd$_2$CoMnO$_6$ also exhibits a larger $H_c$ of about 5 kOe at 100 K. Similar results are also reported in Nd$_2$CoMnO$_6$ prepared by solid-state reaction method and Sm$_2$CoMnO$_6$ [7].

4. Conclusions

Pure La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ powders had been obtained through PVA sol-gel method. XRD analysis
revealed that single phase La$_2$CoMnO$_6$ powders with an average crystallite size of ~20 nm was synthesized through the charring step, while monophasic and well-crystallized Nd$_2$CoMnO$_6$ powders were obtained at 600°C for 2 h. The PVA sol-gel method used in the procedure has the advantages of lower synthesis temperature and shorter preparation time for the synthesis of La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$. Magnetic measurements showed that both La$_2$CoMnO$_6$ and Nd$_2$CoMnO$_6$ powders are paramagnetic at room temperature.

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**Disclosure statement**

No, potential conflict of interest was reported by the authors.

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Figure 11. Magnetic hysteresis loops of Nd$_2$CoMnO$_6$ powders calcined at 600°C for 2 h.

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