Influence of Elastic and Optical Properties on AgFeO_2 and AgCrO_2 Delafossite to be Applied in High-Frequency Applications

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Nanometric AgFeO_2 and AgCrO_2 delafossite were easily prepared by the flash auto-combustion method. The two main bands estimated from FTIR (Fourier-transform infrared) analysis were the tetrahedral A-site (573 cm^{-1} for AgFeO_2 , 630 cm^{-1} for AgCrO_2) and the octahedral B-site (484 cm^{-1} for AgFeO_2 , 595 cm^{-1} for AgCrO_2). This study is mainly focused on the elastic properties evaluated from the FTIR analysis and showed that AgCrO_2 delafossite is more elastic than AgFeO_2 delafossite. The elastic properties can be explained by studying the longitudinal and transverse velocities. Owing to the optical properties results, AgCrO_2 delafossite is a promising material to be applied in optical devices. However, AgFeO_2 delafossite is a promising material in magnetic applications because it showed a large switching field distribution by 9-fold more than that of AgCrO_2 delafossite. Moreover, high-frequency applications were calculated from the magnetic analysis and showed that both samples could be applied in ultra-high microwave applications.

INTRODUCTION

Recently, ABO_2 delafossite-oxides [A is monovalent = silver (Ag), B is trivalent = chromium (Cr) or iron (Fe)] have been paid great attention due to several potential applications, such as water purification, conductors, antimicrobials, chemical sensors, and solar cells.^{1–11} Moreover, it is known that silver (Ag) nanoparticles has high resistance against oxidation and high thermal and electrical conductivity that can be used in a wide range of useful applications.^{12–20} In this work, the study of elastic properties and high-frequency applications is discussed for the first time for AgCrO_2 and AgFeO_2 delafossite; there is no previous literature on this subject. Fourier-transform infrared (FTIR) analysis was determined to study the ordering phenomena of the samples. Moreover, the absorption bands assigned from the FTIR analysis were due to the

oxygen cations and ions vibration at different frequencies.²¹ The elastic constants estimated from the FTIR analysis have a great interest due to their help in understanding the thermal properties of the samples.²¹ Industrialists need to study different properties in detail for different materials to be able to choose the right materials for the right applications. Thus, it is important to illustrate various properties in detail, such as the elastic constants, optical parameters, switching field, and high-frequency responses. It is known that these parameters are strongly dependent on the materials' structure and preparation methods. Many methods are used for preparing nanoparticles, such as ceramic and wet methods.^{22–28} The most effective methods that prepare the nanoparticles with a small size are wet methods, such as citrate, oxalate, flash, sol-gel, and co-precipitation. Flash auto-combustion is used in the present work and has many advantages such as saving time, giving a high yield, and being an effective method. Moreover, flash auto-combustion used to prepare AgFeO_2 and AgCrO_2 delafossite is simple and low-cost, which can have

potential applications in electromagnetic composites.^{29–32} In addition, the characterization of the investigated samples has been discussed in detail in previous literature^{33–36} to assure their formation. Finally, the novelty of the present work is the illustration of the elastic properties estimated from FTIR analysis, as well as the optical properties of AgFeO₂ and AgCrO₂ delafossite. Also, high-frequency applications estimated from the magnetic measurements were studied for both samples. Moreover, to the authors' knowledge, this is the first time that the elastic properties from FTIR analysis and high-frequency applications for the investigated samples have been explained.

EXPERIMENTAL DETAILS

Preparation and Characterization

Figure 1a shows a schematic of the preparation of AgFeO₂ and AgCrO₂ delafossite by the flash method. The starting ingredients were silver, iron III, or chromium III nitrates. They were mixed in stoichiometric ratios after adding a small amount of distilled water. Next, urea was added to the mixture in a stoichiometric ratio. Then, the mixture was heated at 250°C until a fine powder was produced. The powder was ground for half an hour to be ready for analysis. The FTIR analysis was investigated using a Jasco FTIR 300 E spectrometer apparatus. Moreover, the magnetic properties were measured by a vibrating sample magnetometer (Lake Shore 7410). The optical properties were measured using a Jasco-V-570 spectrophotometer (Japan).

RESULTS AND DISCUSSION

FTIR Analysis

Figure 1b and Table I show the FTIR analysis of AgFeO₂ and AgCrO₂ delafossite in the range 400–4000 cm⁻¹. Two main absorption peaks, 1 and 2, appeared for both samples, which indicated the bond vibration of Cr–O or Fe–O in octahedral and tetrahedral sites, respectively. Moreover, peak 3 was assigned to the stretching vibration of water. However, peak 4 appeared due to the C=N group estimated from urea and metal nitrate.^{19,37} Peaks 5 and 6 were assigned to the carboxyl group, while peak 7 was due to the O–H stretching vibration. Finally, peak 8 was strong due to the presence of the NH group and the water molecules. As a result, FTIR analysis assured the formation of AgFeO₂ and AgCrO₂ delafossite.

Elastic Study

The study of the elastic properties was calculated from the FTIR analysis for AgFeO₂ and AgCrO₂ delafossite. It is known that the cation distribution of both samples is important for calculating the force constants.³⁸ The distribution of the cations was as follows:

Moreover, the force constants were calculated as follows:³⁹

$$K_t = 7.62 \times M_A \times v_1^2 \times 10^{-7} \text{N/m} \quad (3)$$

$$K_o = 10.62 \times M_B \times v_2^2 \times 10^{-7} \text{N/m} \quad (4)$$

where K_t is the force constant in the tetrahedral site, K_o is the force constant in the octahedral site, and M_A and M_B are the molecular weights for tetrahedral and octahedral sites, respectively. Tables II and III show the elastic constants for both samples. It can be seen that the values of the force constant at the tetrahedral site (K_t) for both samples were larger than those for the octahedral site (K_o). This was due to the bond length in the tetrahedral structure being shorter than that in the octahedral site.³⁹ It is known that the calculations of the bulk modulus (B), longitudinal elastic velocity (V_l), transverse or shear elastic velocity ($V_t = V_s$), and the mean elastic velocity (V_m) were as follows:^{40–42}

$$B = \frac{1}{3}[C_{11} + 2C_{12}] \quad (5)$$

$$K = aC_{11} \quad (6)$$

$$V_l = \left(\frac{C_{11}}{D_x} \right)^{1/2} \quad (7)$$

$$V_t = \frac{V_l}{\sqrt{3}} \quad (8)$$

$$V_m = \frac{1}{3} \left[\frac{1}{V_l^3} + \frac{2}{V_s^3} \right]^{1/3} \quad (9)$$

where C_{11} and C_{12} are the constants of stiffness, K is the force constant, D_x is the x-ray density, and a is the lattice parameter. In addition to knowing more details about the elasticity of the investigated samples, important parameters were calculated as follows:⁴³

$$L = D_x V_l^2 \quad (10)$$

$$G = D_x V_s^2 \quad (11)$$

$$\sigma = \frac{L - 2G}{2(L - G)} \quad (12)$$

Fig. 1. (a) Schematic of the synthesis of AgCrO₂ and AgFeO₂ delafossite. **b** FTIR spectra of AgCrO₂ and AgFeO₂ delafossite

Table I. The peak values of FTIR analysis of AgCrO ₂ and AgFeO ₂ delafossite								
No. of peaks	1 cm ⁻¹	2 cm ⁻¹	3 cm ⁻¹	4 cm ⁻¹	5 cm ⁻¹	6 cm ⁻¹	7 cm ⁻¹	8 cm ⁻¹
AgFeO ₂	484	573.7	825.4	1359.6	1628.6	1763.6	2396.1	3428.8
AgCrO ₂	555.4	630.6	—	1438.6	1629.6	—	2374.9	3427.9

Table II. The elastic parameters of AgCrO₂ and AgFeO₂ delafossite

Samples	ν_1 tetra $\times 10^2$ (m ⁻¹)	ν_2 octa $\times 10^2$ (m ⁻¹)	M_A $\times 10^{-3}$ (kg)	M_B $\times 10^{-3}$ (kg)	a (Å) ³³⁻³⁶	D_x Kg/ m ³	K_t (N/ m)	K_o (N/ m)	K (N/ m)	ν_{av} $\times 10^2$ (m ⁻¹)	θ_D (K)	C_{11} (Gpa)
AgFeO ₂	573.7	484	81.8566	81.8566	2.888	3950	205.3	203.6	204.5	528.9	12,118.4	707.9
AgCrO ₂	630.6	555.4	107.8682	51.9961	2.988	4460	326.9	170.3	248.6	593.0	13,588.4	831.9

Table III. The elastic parameters of AgCrO₂ and AgFeO₂ delafossite

Samples	V_1 (m/s)	$V_s = V_t$ (m/s)	V_m m/ s	C_{44} (Gpa)	G (Gpa)	L (Gpa)	σ (unitless)	C_{12} (Gpa)	E (Gpa)	B (Gpa)	V_A (m3)	θ'_D (K)
AgFeO ₂	13,388.0	7729.6	8581.3	235.9	235.9	707.9	0.25	132.7	666.1	324.5	12.4	932.4
AgCrO ₂	13,658.1	7885.5	8754.4	277.3	277.3	831.9	0.25	155.9	782.7	381.3	10.8	997.1

$$E = (C_{11} - C_{12})(C_{11} + 2C_{12}) \quad (13)$$

where L is the longitudinal modulus, E is Young's modulus, G is the rigidity modulus, and σ is Poisson's ratio. It can be seen from Tables II and III that these elastic parameters were larger for AgCrO₂ delafossite than for AgFeO₂ delafossite due to the interatomic bonding of the atoms. In addition, the bond strength of the Young's modulus increased in AgCrO₂ delafossite due to the increase of its rigidity. Thus, AgCrO₂ delafossite has more strength than has AgFeO₂ delafossite.

It is known from the theory of isotropic elasticity that Poisson's ratio was in the range from -1 to 0.5. Thus, in the present study, Poisson's ratio is 0.25, which is in good agreement with the theory, indicating that the investigated samples were not brittle.

The calculation of Debye temperature is important to know the conduction mechanism of the investigated sample, and can be calculated from Waldron's equation as follows:³⁸

$$\theta_D = \frac{\hbar c \nu_{av}}{k_B} \quad (14)$$

$$\hbar = \frac{h}{2\pi} \quad (15)$$

$$\nu_{av} = \frac{\nu_t + \nu_o}{2} \quad (16)$$

where h is the Planck constant, c is the velocity of light, ν_{av} is the average frequency, k_B is the Boltzmann constant, and ν_t and ν_o are the tetrahedral and octahedral frequencies, respectively. Table II shows that θ_D is larger for AgCrO₂ delafossite than for AgFeO₂ delafossite due to the larger values of the octahedral and tetrahedral

frequencies of AgCrO₂ compared with AgFeO₂ delafossite. Moreover, Anderson's formula for the Debye temperature calculation θ'_D was as follows:⁴⁴

$$\theta'_D = \frac{h}{k_B} \times \left(\frac{3N_A}{4\pi V_A} \right)^{\frac{1}{3}} \times V_m \quad (17)$$

$$V_A = \left(\frac{M}{D_x} \right) / q \quad (18)$$

where N_A is the Avogadro number, V_A is the mean atomic volume, and q is the number of atoms in the ABO₂ formula (= 4). One can observe from the result of the θ'_D of both samples that the conduction mechanism of AgFeO₂ was due to electrons (n -type).

Optical Properties

Optical properties have attracted attention from researchers because they are very useful in electronic devices.⁴⁵⁻⁴⁷ Figure 2a and Table IV illustrate the refractive index n^2 and the wavelength λ^2 of AgFeO₂ and AgCrO₂ delafossite. Based on the following equation, the lattice dielectric constant (ϵ_l) was obtained as follows:⁴⁸

$$n^2 = \epsilon_l - \left[\frac{e^2}{\pi c^2} \right] \left(\frac{N}{m^*} \right) \lambda^2 \quad (19)$$

where c is the velocity of light, (N/m^*) is the ratio of the free carrier concentration to the effective mass, and e is the electronic charge. It can be seen from Fig. 2a that, at a longer wavelength (λ), the dependence of n^2 versus λ^2 is linear, and the dielectric constant (ϵ_l) is estimated from the extrapolation of the linear part to zero λ . Moreover, the values of ($e^2 N / \pi c^2 m^*$) were also estimated from this linear part and are reported in Table IV. Generally, the increase of the charge carriers decreases the crystallinity of the material.^{49,50}

Fig. 2. **a** The refractive index n^2 versus the wavelength λ^2 of AgCrO_2 and AgFeO_2 delafossite. **b** The refractive index $(n^2 - 1)^{-1}$ versus the photon energy $(h\nu)^2$ of AgCrO_2 and AgFeO_2 delafossite. **c** The refractive index $(n^2 - 1)^{-1}$ versus the wavelength λ^{-2} of AgCrO_2 and AgFeO_2 delafossite

Table IV. The operating frequency and the optical parameters of AgCrO₂ and AgFeO₂ delafossite

Samples	ω (GHz)	E_o (eV)	E_d (eV)	ϵ_1	$(e^2 N / \pi c^2 m^*) \times 10^{-7}$ (kg ⁻¹ m ⁻³)	$\chi^{(3)}$ (esu) $\times 10^{-11}$	n_∞	λ_o (nm)	S_o (nm ⁻²)
AgFeO ₂	2.49	6.09	34.20168	5.37	6.27	1.19	2.57	0.2038	135.1
AgCrO ₂	0.42	8.96	62.00544	6.84	5.71	6.52	2.81	0.1392	357.1

Thus, AgCrO₂ is more crystalline than AgFeO₂ delafossite due to the (N/m^{*}) of the AgCrO₂ delafossite being smaller than that of AgFeO₂ delafossite.

Figure 2b shows the relationship between $(n^2 - 1)^{-1}$ versus $(h\nu)^2$ for AgFeO₂ and AgCrO₂ delafossite. At low optical frequencies, the dispersion parameters (E_o and E_d) of the investigated samples can be obtained from the following equation:⁵¹

$$(n^2 - 1)^{-1} = -\frac{1}{E_o E_d} (h\nu)^2 + \frac{E_o}{E_d} \quad (20)$$

where E_o is the single oscillator energy and E_d is the dispersion energy. Table IV shows the values of E_d and E_o estimated from the intercept of the linear part of the graph, and they give information about the measure of the strength of the interband optical transition and the average excitation for the electronic transition, respectively.

Moreover, Fig. 2c shows the variation of $(n^2 - 1)^{-1}$ versus λ^{-2} of AgCrO₂ and AgFeO₂ delafossite and can be expressed from the following relationship:⁵²

$$\frac{n_\infty^2 - 1}{n^2 - 1} = 1 - \left(\frac{\lambda_o}{\lambda}\right)^2 \quad (21)$$

where λ_o is the average wavelength of the interband oscillator and n_∞ is the refractive index of the long wavelength. The average oscillator strength, S_o , was estimated from the slope and the intercept of the graph of Fig. 2c. Based on these results, all these optical parameters may help in tailoring the construction of optical devices.

Figure 3a shows the variation of the third-order nonlinear optical susceptibility versus the photon energy ($h\nu$) for AgCrO₂ and AgFeO₂ delafossite. The equation of the third-order nonlinear susceptibility, $\chi^{(3)}$, was expressed as follows:⁵³

$$\chi^{(3)} = A = \left[\frac{E_o E_d}{4\pi (E_o^2 - (h\nu)^2)} \right]^4 \quad (22)$$

where A is 1.7×10^{-10} . It can be seen from Fig. 3a and Table IV that $\chi^{(3)}$ of AgCrO₂ delafossite is higher than that of AgFeO₂. As a result, AgCrO₂ delafossite is a more promising material for applying in optical devices than AgFeO₂ delafossite.

Fig. 3. **a** The third-order nonlinear susceptibility versus the photon energy of AgCrO₂ and AgFeO₂ delafossite. **b** The switching field distribution of AgCrO₂ and AgFeO₂ delafossite

Switching Field Distribution and High-Frequency Applications

Figure 3b shows the switching field distribution estimated from the first derivative of the magnetization of AgFeO₂ and AgCrO₂ delafossite. The magnetic properties of the investigated samples have been illustrated in detail in previous studies.^{33–36} It can be seen that AgFeO₂ delafossite had a strong switching field distribution more than 9-fold that of AgCrO₂ delafossite. This may be due to the strong magnetic interaction of the AgFeO₂

Fig. 4. The operating frequency versus the applied field of AgCrO₂ and AgFeO₂ delafossite

cations. As a result, AgFeO₂ delafossite is a promising material to be applied in magnetic applications.

Figure 4 and Table IV show the high microwave operating frequency response of the investigated samples. It is important to calculate the microwave operating frequency to use a certain frequency band to avoid the interference of the frequency.⁵⁴ The microwave operating frequency was calculated from the following relationship:⁵⁵

$$\omega = 8\pi^2\gamma M \quad (23)$$

where $\gamma = 2.8$ (MHz/G) which is the gyromagnetic ratio and M is the magnetization of AgFeO₂ and AgCrO₂ delafossite. It can be seen that the operating frequency of AgFeO₂ delafossite is larger by 5-fold than that of AgCrO₂ delafossite, as shown in

Table IV. This means that AgFeO₂ could be applied in the microwave ultra-high frequency *L* band because it has the ω at 2.49 GHz. However, the AgCrO₂ delafossite could be applied in the microwave ultrahigh-frequency *P* band because it has the ω at 420 MHz.

CONCLUSION

AgFeO₂ and AgCrO₂ delafossite were successfully prepared by the flash auto-combustion method. The main peaks of the FTIR analysis assured the formation of the investigated samples. In addition, the elastic properties were estimated from the FTIR analysis and showed that AgCrO₂ delafossite has more strength than AgFeO₂ delafossite. The optical properties were studied and showed that AgCrO₂

delafossite is a more promising material for applying in optical devices. In addition, the switching field distribution was studied and showed that AgFeO₂ is larger by 9-fold than that of AgCrO₂ delafossite. Thus, AgFeO₂ delafossite is a promising material in magnetic applications. Moreover, high-frequency applications were studied by magnetic analysis and showed that AgFeO₂ could be applied in the microwave ultra-high frequency L band and AgCrO₂ delafossite could be applied in the microwave ultrahigh-frequency P band. For the future, the authors suggest improving this work by studying the investigated samples using citrate or sol-gel methods.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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