Unique edge-sharing sulfate-transition metal coordination in $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co)

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Two new compounds, $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ and $\text{Na}_2\text{Co}(\text{SO}_4)_2$, were synthesized and their structure and properties were characterized. They form a new structure type, containing a bidentate coordination of sulfate to the transition metal center, which was determined via single crystal X-ray diffraction combined with model refinements to both laboratory X-ray and time-of-flight neutron powder diffraction data. The compounds were both found to crystallize in the $C2/c$ space group with $Z = 24$ and a unit cell of $a=23.3461(3)$ Å, $b=10.3004(1)$ Å, $c=17.4115(2)$ Å, $\beta=98.8659(9)^\circ$, and $V=4136.99(8)$ Å³ for the cobalt analog and $a=23.2253(1)$ Å, $b=10.26155(6)$ Å, $c=17.3353(1)$ Å, $\beta=99.0376(5)^\circ$, and $V=4080.20(5)$ Å³ for the nickel analog. Magnetization measurements show that the transition metal centers have negligible interactions with neighboring sites. Infrared and Raman spectroscopies were used to further probe the unique

sulfate-transition metal coordination, and confirm the bidentate binding motif. The resulting pseudo-trigonal bipyramidal coordination produces vivid violet, $\text{Na}_2\text{Co}(\text{SO}_4)_2$, and yellow, $\text{Na}_2\text{Ni}(\text{SO}_4)_2$, colors that were probed by diffuse reflectance.

Keywords: Sulfate, coordination, X-ray diffraction, battery, pigment

1. Introduction

Energy storage may support future renewable technology development by improving grid reliability and lowering overall system costs [1]. Solid state materials are promising battery materials that may meet the need for energy storage due to their ability to store and transport alkali ions. In addition to three-dimensional framework materials containing phosphates and silicates, three-dimensional sulfate materials have been studied as battery materials due to their structural stability and compositional flexibility [2]. One of the early sulfate materials to be studied for its lithium ion insertion capabilities, $\text{Fe}_2(\text{SO}_4)_3$, was determined to be a superior ionic conductor, possessing a larger open circuit voltage relative to $\text{Fe}_2(\text{WO}_4)_2$ due to the increased covalency between S-O relative to W-O which ultimately decreases the Fe-O covalency in the Fe-O-S linkage [3]. Furthermore, the presence of channels in many three dimensional framework materials provides a means for the insertion of alkali ions while maintaining the structural stability due to the three-dimensional network of connected polyhedra that does not collapse or expand to as large an extent as do layered compounds [2,4].

In this study, we present the structure and physical properties of two isomorphous compounds, $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ and $\text{Na}_2\text{Co}(\text{SO}_4)_2$, which could be of interest for battery materials given the presence of both electron withdrawing sulfate groups and open channels to promote the movement of sodium ions. Through single crystal X-ray diffraction, laboratory powder X-ray

diffraction, and time-of-flight (TOF) neutron powder diffraction, we identify a bidentate binding motif of the sulfate tetrahedra to the transition metal ions. The unique edge-sharing coordination between the octahedra and half of the sulfate tetrahedra was further confirmed by Infrared and Raman spectroscopy. Curie-Weiss fits of temperature-dependent magnetization data show that both compounds are paramagnetic with negligible interactions between transition metal sites. The highly localized nature of the transition metal d electrons combined with the bidentate sulfate binding motif gives rise to $d-d$ transitions resulting in vibrant violet (cobalt analog) and yellow (nickel analog) colors. The lack of inter-site transition metal interactions implies that the electrical conductivity of these materials will likely remain low, requiring significant nanostructuring to become a viable battery cathode. However, the strong colors that result from the unique sulfate-transition metal coordination may find use in pigments and related applications.

2. Materials and methods

2.1. Synthesis

Initial syntheses were carried out targeting the composition $\text{NaNi}_2(\text{SO}_4)_2\text{Cl}$ which is reported to exist, but for which no structure solution exists [5]. However, X-ray diffraction consistently showed the presence of nickel chloride and/or nickel oxide in the products in addition to the target phase, which only went away when the target formula was adjusted to $\text{Na}_2\text{Ni}(\text{SO}_4)_2$. With knowledge of the correct formula, it was possible to synthesize single phase polycrystalline samples of $\text{Na}_2M(\text{SO}_4)_2$ ($M=\text{Ni}$ and Co) via traditional solid state synthesis. The reagents, Na_2SO_4 (Mallinckrodt, 99.3%), $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ (Alfa Aesar 99.97%), and $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$ (Acros Organics 99+%), were ground in agate mortar and pestle, pressed into pellets, and placed

in uncovered alumina crucibles. Samples were heated at 580 °C for 72 hr with a 50 °C/hr ramp rate. Single crystals of $\text{Na}_2\text{Co}(\text{SO}_4)_2$ were prepared by first making the polycrystalline sample under argon at 550 °C for 71 hours, followed by a regrinding and heating in air in an alumina crucible at 600 °C for up to 10 minutes. The sample was then melted at 700 °C for one minute and quickly removed from the furnace and poured onto a stainless steel plate to re-solidify. Polycrystalline precursors can be accessed in argon or air. A micron-sized single crystal suitable for X-ray diffraction was selected from the recrystallized boule.

2.2. Diffraction

Single crystal X-ray diffraction (XRD) data of $\text{Na}_2\text{Co}(\text{SO}_4)_2$ were collected by mounting a small single crystal on a loop with a minimum of Paratone-N oil. The temperature of the crystal was then fixed at $T = 110(2)$ K using a Cryojet system (manufactured by Oxford Instruments). All reflection intensities were measured using a Xcalibur3 (equipped with a Sapphire3 CCD) employing Mo $K\alpha$ radiation ($\lambda = 0.71073$ Å) under the CrysAlisPro software package (version 1.171.36.32, Agilent Technologies, 2013). Unit cell indexing and data reductions were performed using the CrysAlisPro software. The generation of the initial models and structure refinements were performed using SIR97 [6] and SHELXL-2013 [7], respectively. The selection of the $C2/c$ (No. 15) was based on the observed Laue symmetry $2/m$ and the systematic absences. The collected data were corrected for absorption using a multifaceted crystal model based on expressions derived by R.C. Clark & J.S. Reid (analytical numeric absorption correction) [8].

Both compounds were probed with laboratory powder XRD data were collected on a Bruker D8 Focus (40 kV, 40 mA, sealed Cu X-ray tube, $K_{\alpha 1} = 1.540596$ Å, $K_{\alpha 2} = 1.544493$ Å ,

ratio of $K_{a2}/K_{a1}=0.529480$) equipped with Ni filter and a LynxEye detector. TOF neutron diffraction data was collected on both compounds at the High-Intensity Powder Diffractometer (HIPD) at Los Alamos National Laboratory's Lujan Neutron Scattering Center. Neutron data was collected at $T = 300$ K and $T = 15$ K, however no structural change was observed beyond the expected lattice contraction upon cooling. Therefore, all eight banks of the $T = 300$ K neutron data were used in combined refinements with the room temperature laboratory powder XRD using the GSAS software package with the EXPGUI user interface [9,10].

2.3. Magnetism

Magnetization data for $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co) were collected using the ACMS option of a 9-Tesla Quantum Design Physical Properties Measurement System (PPMS). The temperature-dependent magnetization data for both compounds were obtained under zero-field cooled (ZFC) and field-cooled (FC) conditions from $T = 1.8$ K – 300 K under an applied field of $\mu_0 H = 5$ T.

2.4. Spectroscopy

Diffuse reflectance data were collected on an Ocean Optics ultraviolet-visible spectrometer equipped with a Microprobe deuterium-halogen light source. The reflectance data, R , were transformed to approximate reflectance, $F(R)$ using the Kubelka-Munk function,

$$F(R) = \frac{(1-R)^2}{2R}.$$

The infrared (IR) spectra were collected on a Bruker Vector 33 FT-IR spectrophotometer with an ATR attachment. Raman data was collected on Jobin-Yvon T64000 Raman spectrometer, equipped with an Olympus microscope and a single monochromator option. The 514.5 nm line of an Ar⁺ gas Coherent laser was used for excitation.

3. Results and Discussion

Single crystal XRD of $\text{Na}_2\text{Co}(\text{SO}_4)$ was used to determine the unit cell, metal positions, and approximate sulfur and oxygen positions (not given). The presence of *C*-centering, rather than a smaller primitive cell masquerading as a larger *C*-centered due to crystal twinning that could be present in the single crystal, was confirmed by Le Bail fits of the laboratory powder XRD data. The observed reflections could be well-indexed by a cell with space group *C2/c*, $a \approx 23.35 \text{ \AA}$, $b \approx 10.30 \text{ \AA}$, $c \approx 17.41 \text{ \AA}$, and $\beta \approx 98.87^\circ$, but not with a smaller primitive cell. Final unit cells for both the cobalt and nickel compounds are given in Table 1.

Due to the low scattering power of oxygen relative to the other elements, and previous mis-assignments of sulfate coordinations based solely on X-ray data [11,12,13], neutron diffraction data is needed to accurately determine the oxygen positions and concomitant sulfur coordination of the metal atoms. TOF neutron diffraction data and powder XRD data were combined in a single refinement to increase the number of observables. The model obtained from single crystal data was used as a starting model for the refinement. After refinement of the sodium and transition metal atom positions, sulfate tetrahedral rigid body constraints, with a S-O bond length of $1.5 \text{ \AA}$, were used to establish regular tetrahedra centered at the single crystal XRD sulfur positions. The rigid bodies were allowed to rotate to determine the coordination of the transition metal to the sulfate tetrahedra. After convergence, the rigid bodies were removed and replaced with a S-O distance restraint of $1.5 \pm 0.05 \text{ \AA}$ with a restraint weight of 25. The refined fits to the $\text{Na}_2\text{Co}(\text{SO}_4)_2$ data are shown in Figure 1 and the refined fits to the $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ data are presented in the supporting information. To check the uniqueness of the crystallographic solution, the single crystal data was re-refined using the same structural model as the one refined from powder diffraction data, but all symmetry constraints were removed. The structure was

allowed to converge and a similar structure was obtained. Final crystallographic information from the joint neutron X-ray refinements is shown in Table 1. Full atomic positions and displacement parameters are reported in the crystallographic information files in the Supporting Information.

Figure 1. Laboratory X-ray diffraction, room temperature, and TOF neutron diffraction, 300 K, (denoted as Bank 1-8) patterns for $\text{Na}_2\text{Co}(\text{SO}_4)_2$: data (black), Rietveld refinement fit (red), difference curve (gray), and tick marks denote allowed peak positions $\text{Na}_2\text{Co}(\text{SO}_4)_2$ (blue) and Na_2SO_4 (green). Pairs of banks (top and bottom) share the same d -spacing as labeled at the bottom of the figure. Structure shown in upper left hand corner: transition metal octahedra (blue), sulfate tetrahedra (yellow), sodium atom (green), and oxygen (red).

The isomorphic structure of these two compounds is shown in Figure 2. The transition metal octahedra are arranged as pairs, as depicted in Figure 2a, that are bridged by two sulfate tetrahedra; half of the sulfate tetrahedra bridge two transition metal octahedra and are also edge-sharing with the transition metal octahedra the other half of the tetrahedra only share

corners with the octahedra to form a polyhedra network. As a result of this unique coordination each transition metal octahedron is edge-sharing with one sulfate tetrahedron and corner-sharing with four other sulfates. The bond lengths for the transition metal octahedra are shown in Table 2. The tetrahedra and octahedra form a polyhedra network, that when viewed in the *ac* plane with a 30° rotation around *b* reveals open channels that house the sodium ions. The empty channels are shown in Figure 2b and the full structure, including the sodium ions, is shown in Figure 2c. These channels provide a pore that could aid in the mobility of the sodium ions which is of interest for potential battery materials.

The most surprising structural characteristic is the unique bidentate (edge-sharing) coordination between the sulfate tetrahedra and the transition metal octahedra. Sulfate edge-sharing coordination to individual ions has previously only been seen in a few other compounds, such as Na₂SO₄ [14] and CsPr(SO₄)₂ [15], but is usually associated with a metal site coordination number greater than six (cf. eight for Pr in CsPr(SO₄)₂) and has never before, to our knowledge, been seen with a transition metal cation. As required by the rigid O-S-O tetrahedral bond angle, the transition metal-oxygen bond lengths are longer for the bidentate coordination motif (highlighted in blue Table 2, although they are not as elongated as in Na₂SO₄ [14] and CsPr(SO₄)₂ [15]). This shows that there is not a relatively large transition metal displacement to accommodate the edge-sharing sulfate. Additionally, Table 2 contains the bond valence sums, $\nu_{ij} = \exp\left[\frac{R_0 - d_{ij}}{0.37}\right]$, and the distortion index, $D = \frac{1}{n} \sum_n \frac{|d_{ij} - d_{avg}|}{d_{avg}}$, where R_0 is the bond valence parameter, d_{ij} is the bond length between atoms *i* and *j*, d_{avg} is the average bond length in the polyhedra, and *n* is the number of bonds defining the polyhedra. The bond valence sums for both analogs are in reasonable agreement with an expected valence of two, which indicates that this is a reasonable structure with respect to the transition metal coordination. The distortion index,

being slightly higher for the cobalt analog than the nickel analog, indicates that on average the octahedra of the cobalt analog are slightly more distorted than the nickel analog.

Figure 2. a) Two blue distorted transition metal octahedra (oxygen in red) bridged by two sulfate tetrahedra are shown here. Each bridging sulfate tetrahedra shares an edge with one octahedron and a corner with the other. All of the remaining corners of the octahedra are corner sharing with four tetrahedra forming a polyhedral network. The network formed by the octahedra and tetrahedra are shown in b) and the full structure containing the sodium atoms (green) is shown in c).

Table 1. Crystallographic data obtained from combined laboratory powder XRD and neutron refinements of Na₂Co(SO₄)₂ and Na₂Ni(SO₄)₂^{aa}

	Na ₂ Co(SO ₄) ₂	Na ₂ Ni(SO ₄) ₂
space group	<i>C2/c</i> (15)	<i>C2/c</i> (15)
<i>a</i> (Å)	23.3461(3)	23.2253(1)
<i>b</i> (Å)	10.3004(1)	10.26155(6)
<i>c</i> (Å)	17.4115(2)	17.3353(1)
β (deg)	98.8659(9)	99.0376(5)
<i>V</i> (Å ³)	4136.99(8)	4080.20(5)
<i>Z</i>	24	24
formula weight (g/mol)	7128.60	7123.01
calculated density (g/cm ³)	2.86	2.90
<i>T</i> (K) (neutron/XRPD)	300/RT	300/RT
number of reflections	48259	48241
χ^2	1.487	1.968
<i>R</i> _{wp}	1.81	2.39
<i>R</i> _p	1.33	1.88

^a The goodness of fit parameters are defined as follows:

$$\chi^2 = \frac{M}{(N_{obs} - N_{var})}, R_{wp} = \sqrt{\frac{\sum w(I_o - I_c)^2}{\sum w I_o^2}}, \text{ and } R_p = \frac{\sum |I_o - I_c|}{\sum I_o}$$

Table 2. Bond lengths, bond valence sums, and distortion index of transition metal cations obtained from combined laboratory powder XRD and neutron refinements

Na ₂ Co(SO ₄) ₂				Na ₂ Ni(SO ₄) ₂			
Bond	Multiplicity	Bond Lengths (Å)	v _{ij}	Bond	Multiplicity	Bond Lengths (Å)	v _{ij}
Co1-O18	×1	1.944(7)	0.507	Ni1-O18	×1	1.918(4)	0.519
Co1-O17	×1	2.073(6)	0.357	Ni1-O11	×1	2.041(4)	0.372
Co1-O11	×1	2.095(7)	0.336	Ni1-O17	×1	2.077(4)	0.337
Co1-O6	×1	2.221(7)	0.240	Ni1-O6	×1	2.098(4)	0.319
Co1-O21	×1	2.165(7)	0.279	Ni1-O4	×1	2.140(4)	0.284
Co1-O4	×1	2.192(8)	0.259	Ni1-O21	×1	2.264(4)	0.204
		Σv _{ij}	1.977			Σv _{ij}	2.036
		D	0.037			D	0.037
Co2-O10	×1	1.802(7)	0.744	Ni2-O10	×1	1.951(4)	0.474
Co2-O1	×1	1.970(9)	0.472	Ni2-O7	×1	1.987(4)	0.431
Co2-O7	×1	2.088(7)	0.343	Ni2-O1	×1	2.081(4)	0.333
Co2-O20	×1	2.169(7)	0.276	Ni2-O2	×1	2.082(4)	0.333
Co2-O3	×1	2.232(8)	0.232	Ni2-O20	×1	2.122(4)	0.299
Co2-O2	×1	2.299(7)	0.194	Ni2-O3	×1	2.136(4)	0.288
		Σv _{ij}	2.260			Σv _{ij}	2.158
		D	0.067			D	0.029
Co3-O12	×1	1.974(7)	0.466	Ni3-O24	×1	2.004(5)	0.411
Co3-O24	×1	2.013(7)	0.420	Ni3-O12	×1	2.041(4)	0.372
Co3-O5	×1	2.076(7)	0.354	Ni3-O13	×1	2.048(5)	0.365
Co3-O8	×1	2.158(7)	0.284	Ni3-O23	×1	2.128(4)	0.294
Co3-O23	×1	2.298(7)	0.194	Ni3-O8	×1	2.114(4)	0.305
Co3-O13	×1	2.192(7)	0.259	Ni3-O5	×1	2.153(4)	0.274
		Σv _{ij}	1.977			Σv _{ij}	2.022
		D	0.046			D	0.024

To further corroborate the unique sulfate coordination, several spectroscopic studies were performed. Both IR and Raman spectroscopy are sensitive to the coordination of the sulfate ion. It is well known that as the sulfate ion coordinates to one or two ligands the symmetry of the ion goes from T_d for the free ion, to C_{3v} for the unidentate ion, and C_{2v} for the bidentate ion [16]. Lowering of symmetry splits the normal stretching modes of the tetrahedron, shown in Figure 3, with the ν₃ stretch being particularly sensitive to coordination changes (Table 3).

Figure 3. Normal modes of a sulfate tetrahedron

Table 3. Normal mode decomposition of the ν_3 mode of a tetrahedron based on decrease in symmetry

Coordination	Symmetry	Changes in ν_3 mode		
Free	T_d	$\nu_3 (T_2)$		
Unidentate	C_{3v}	$\nu_{3a}(A_1)$	$\nu_{3b}(E)$	
Bidentate	C_{2v}	$\nu_{3a}(A_1)$	$\nu_{3b}(B_1)$	$\nu_{3c}(B_2)$

The splitting of the ν_3 mode has previously been observed in both solution phase systems as well as solid state compounds [17]. Edge-sharing tetrahedra cause two of the ligated oxygens to be very similar in energy and thus can be thought of as bidentate. This is directly observed in the IR, Figure 4a, as a splitting into three bands of the ν_3 mode relative to the free sulfate ion in the reference compound $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$, as predicted by the mode decomposition. The additional broadening is due to the presence of multiple crystallographically independent sulfate anions. In the Raman data, Figure 4b, the ν_3 mode disappears or moves to lower wavenumbers ($\text{Na}_2\text{Ni}(\text{SO}_4)_2$) when the sulfate ion is edge-sharing as in the target compounds, relative to the free ion in Na_2SO_4 . This is due to the splitting and broadening of this mode as observed in the IR. Several additional modes appear in the Raman data that can be attributed to additional non-intra-sulfate stretching modes.

Figure 4. Spectroscopic probes of the sulfate tetrahedra coordination IR and Raman are shown in a) and b) respectively. Kubelka-Munk transformed diffuse reflectance data shown in c) corroborates the observed colors of the compounds shown in d) which is a result of the loss of inversion center and thus allowed *d-d* transitions.

Diffuse reflectance was used to further probe the coordination of the transition metal ion as it is sensitive to the *d-d* transitions. Photographs of the compounds (Figure 4d) show the observed color which is in agreement with the Kubelka-Munk transformed diffuse reflectance spectra for both compounds in Figure 4c. Both compounds show one main absorption peak; $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ absorbs ~390-500 nm, resulting in a yellow color, and $\text{Na}_2\text{Co}(\text{SO}_4)_2$ absorbs ~440-620 nm, resulting in a violet color. The cobalt analog possesses a broader asymmetric peak than the nickel analog. This would be expected due to the cobalt analog possessing a higher degree of distortion than the nickel analog. In both cases, the strong absorption indicates a significant deviation from pure octahedral coordination in which *d-d* transitions are strictly orbitally forbidden. This is a direct consequence of the bidentate coordination by sulfate: the rigid O-S-O tetrahedral bond angle forces the transition metal-oxygen bond lengths for the bidentate coordination motif to be elongated. The result is a distortion that, geometrically, acts as though the material were heading toward trigonal bipyramidal coordination with the bidentate ligand replacing one of the equatorial binding positions (Figure 4d). This breaks the inversion center around the transition metal center, making *d-d* transitions orbitally allowed, as observed. Thus these structures might provide another route to high performance pigments based on trigonal bipyramidal-type coordinations [18].

Figure 5. The inverse of $\chi - \chi_0$ vs. temperature (T) for $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ (gold) and $\text{Na}_2\text{Co}(\text{SO}_4)_2$ (purple). The magnetization data was collected under zero-field cooled (ZFC) conditions in an applied field of 5 Tesla ($\mu_0 H = 5$ T).

Figure 5 shows the temperature-dependent inverse magnetic susceptibility (χ) corrected for a temperature independent contribution (χ_0) vs. T , where magnetization data was collected under zero-field cooled (ZFC) conditions in an applied field of 5 Tesla ($\mu_0 H = 5$ T). The susceptibility curves for $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co) collected under field cooled conditions (not shown) are comparable. No discontinuities or cusps, which might correspond to long range magnetic order, are observed in the data, suggesting that $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co) remains paramagnetic down to the lowest temperature measured ($T \sim 1.8$ K).

Linear fits (black lines) were performed to the inverse ($\chi - \chi_0$) between temperatures of 15 K-300 K for $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co). This model fits accurately reproduce the data showing that both $\text{Na}_2M(\text{SO}_4)_2$ ($M = \text{Ni}$ and Co) are Curie-Weiss like. The magnetic moments determined from fits to the data for $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ and $\text{Na}_2\text{Co}(\text{SO}_4)_2$ are $\mu_{\text{eff}} = 3.23(1) \mu_B$ and

$\mu_{\text{eff}} = 4.92(9) \mu_{\text{B}}$, respectively. These values are in between those for spin-only and spin-orbital contributions calculated using Hund's rules and nicely agree with the μ_{exp} values ($\text{Ni}^{2+} = 3.12 \mu_{\text{B}}$ and $\text{Co}^{2+} = 4.90 \mu_{\text{B}}$) [19] determined empirically. The Weiss temperatures (θ_{w}) were calculated to be $-1.14(8) \text{ K}$ and $-10.6(7) \text{ K}$ for $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ and $\text{Na}_2\text{Co}(\text{SO}_4)_2$, respectively, suggesting that both compounds contain small antiferromagnetic correlations. The larger θ_{w} for $\text{Na}_2\text{Co}(\text{SO}_4)_2$ compared to $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ is likely due to greater dipolar interactions. In both cases, the small values of the Weiss temperatures combined with the lack of apparent magnetic ordering indicate that the interactions between adjacent metal centers are small. This is consistent with the structure, which can be viewed as isolated transition metal octahedra. Such weak interactions, however, do suggest that significant nanostructuring and embedding in a conducting matrix will be required to be viable in sodium battery cathode applications.

4. Conclusions

The compounds $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ and $\text{Na}_2\text{Co}(\text{SO}_4)_2$ have been synthesized and characterized structurally using both diffraction and spectroscopic techniques. The unique structure required the use of single crystal X-ray diffraction combined with powder laboratory X-ray diffraction and neutron diffraction. Further corroboration of the structure was gleaned from infrared and Raman spectroscopies. The vibrant colors were probed via diffuse reflectance spectroscopy and magnetization measurements were run to probe the magnetic moments. The structure of these materials makes them good candidates for further studies into battery applications and for their potential use as pigments due to the strong inversion symmetry breaking at the transition metal site.