Structure and Physical Properties of (x-fluoroanilinium) (benzo[18]crown-6)[FeCr(oxalate)$_3$] Crystals

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The development of multiferroic materials is one of the most attractive topics because of the scientific significance of the coexistence of magnetic ordering and ferroelectricity and the potential for applications in data storage and signal processing. However, multiferroic materials are very rare examples, and the most studied are inorganic transition metal oxides. On the other hand, materials containing organic materials can introduce features unique to inorganic materials, such as designability of molecular structures, sensitive response to microstimuli, and control of dimensionality and anisotropy based on anisotropic molecular structures. Our group have found that (*x*-FAni$^+$)(benzo[18]crown-6)[Mn^{II}Cr^{III}(ox)$_3$]$^-$ (*x*- = *o*-, *m*- and *p*-, FAni = fluoroanilinium, and ox = oxalate) are promising candidates for multiferroic materials. In this study, in order to investigate the effect of metal substitution on the structure and physical properties, we have synthesized and evaluated the structure and magnetic properties of the Fe^{II} analogues, (*x*-FAni$^+$)(benzo[18]crown-6)[Fe^{II}Cr^{III}(ox)$_3$]$^-$ (*x*- = *o*- (**1**), *m*- (**2**) and *p*- (**3**)).

Crystals **1**, **2**, and **3** were synthesized by the solvent diffusion method. The crystal structure of **1** is shown in Fig. 1. The Fe^{II} and Cr^{III} ions were six coordinated by ox^{2-} to form a two-dimensional (2D) honeycomb layered structure. Supramolecular cations arranged between the [Fe^{II}Cr^{III}(ox)$_3$]$^-$ layers. The 2D layered structures consisting of anions and cations, respectively were arranged alternately along the *a*-axis. Alternate stacking structures were also observed in crystal 2 and 3. Temperature product of magnetic susceptibility (χ_m) increased with decreasing temperature and reached the maximum at 5, 14, 13 K for **1**, **2**, and **3**, respectively, indicating ferromagnetic properties of the salts. The magnetization curves at 1.8 K showed hysteresis but is not saturated at 5 T, indicating the canted ferromagnetism of the crystals **1**, **2**, and **3**. Dielectric properties of the crystals will be discussed.

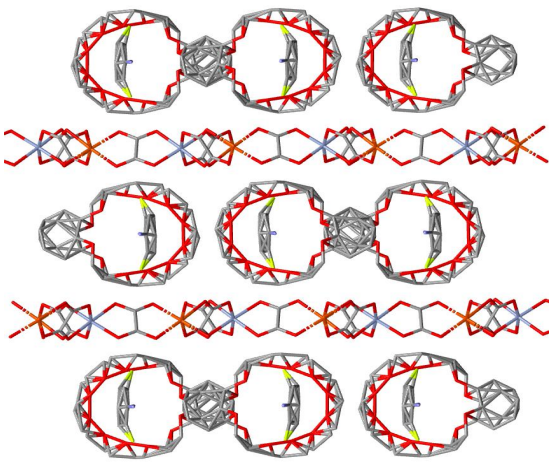

Fig.1. Packing structures of **1** viewed along the *a*-axis