

Lead-free hybrid ferroelectric material based on formamidine: $[\text{NH}_2\text{CHNH}_2]_3\text{Bi}_2\text{I}_9$.

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The extraordinary thermal stability of $[\text{NH}_2\text{CHNH}_2]_3\text{Bi}_2\text{I}_9$ ($\text{FA}_3\text{Bi}_2\text{I}_9$) crystals, up to around 600 K, has been enhanced by using the evaporating method. DSC measurements have revealed reversible structural phase transitions: at 203 K (Phase I \rightarrow Phase II), 173 K (II \rightarrow III), 134.7 K (III \rightarrow IV), 131.4 K (IV \rightarrow V), and 120 K (V \rightarrow VI). The crystal structures of $\text{FA}_3\text{Bi}_2\text{I}_9$ adopt the polar space groups $P6_3mc$ at 220 K (Phase I) and $Cmc2_1$ at 190 K (Phase II). The pyroelectric current (I_{pyr}) measured in the temperature range covering all the phases of $\text{FA}_3\text{Bi}_2\text{I}_9$ confirmed their polar nature. The reversibility of the spontaneous polarization in each phase has been confirmed by the observed hysteresis loops. All the phase transitions are dielectrically active. The dielectric response ($\epsilon^*(\omega, T)$ close to 134.7 and 131.4 K) is characteristic of ferroelectrics with a critical slowing-down process. Optical bandgaps estimated from the UV-vis measurements and calculated using the DFT method are equal to 1.85 and 1.89 eV, respectively. On the basis of the structural, dielectric, and spectroscopic results, the molecular mechanisms of the phase transitions have been proposed.

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