

## Unprecedented solid-state chemical reaction—from $(C_3N_2H_5)_3SbBr_6 \cdot H_2O$ to $(C_3N_2H_5)_5Sb_2Br_{11}$ . From centrosymmetric to non-centrosymmetric crystal structure.

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Tris(imidazolium) hexabromoantimonate(III) hydrate,  $(C_3N_2H_5)_3SbBr_6 \cdot H_2O$ , (abbreviated as TIBA) has been synthesized and characterized by X-ray (at 295, 225, 160, and 110K), differential scanning calorimetry, dilatometry, and dielectric spectroscopy. At room temperature (phase I), the structure consists of discrete  $SbB_6^-$  anions, disordered imidazolium cations, and water molecules forming a 3D array of hydrogen bonds. Below room temperature, TIBA was found to undergo isostructural discontinuous phase transition at 212/221K (cooling–heating) ( $P21/c \leftrightarrow P21/c$ ). The phase transition mechanism is characterized by two contributions: an order–disorder (cationic substructure) and a displacive (water molecules) one. At high temperatures, the TIBA crystal was found to undergo an unprecedented in situ solid-state chemical reaction:



$(C_3N_2H_5)Br(S) + 2H_2O$  This chemical transformation leads to multiphase crystallites dominated by an amorphous phase of  $(C_3N_2H_5)_5Sb_2Br_{11}$ . The creation of ferroelectric crystallites –  $(C_3N_2H_5)_5Sb_2Br_{11}$  – in an “annealed” sample of  $(C_3N_2H_5)_3SbBr_6 \cdot H_2O$  was confirmed by X-ray diffraction phase analysis, dielectric spectroscopy, and pyroelectric measurements. The dielectric response of the electric permittivity and the critical slowing down of the process observed near 140K in the “annealed” sample of TIBA are treated as a “fingerprint” of a neat  $(C_3N_2H_5)_5Sb_2Br_{11}$  ferroelectric.

### Słowa kluczowe

Halogenoantimonates(III), phase transition, hydrogen bonds, Ferroelectric hybridmaterials

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