Organic-inorganic hybrid ferroelectrics (OIHFs) comprise of inorganic frameworks and organic moieties. These materials possess the advantages of the lightweight, structural flexible and environmental friendly features from the organic moiety, and the mechanically strong and robust electro-optical properties from the inorganic backbone. Recently, research on the piezoelectric and electrostrictive properties of OIHFs has been progressed significantly. The piezoelectric coefficients of some OIHFs even exceed those of the benchmark oxide perovskites ferroelectrics such as bismuth barium titanate and lead zirconate titanate. However, the origins of these superior piezoelectric responses are still not clear. In this thesis, I report the discovery and mechanism analysis of a new OIH which produces colossal ferroelastic shear strain about two orders of magnitude higher than that of typical oxide piezoelectrics and giant shear piezoelectric response that is comparable to top-performance Pb-based relaxor.