Piezoelectric materials convert between electrical and mechanical energies such that an applied stress induces a polarization and an applied electric field induces a strain. This review describes the fundamental mechanisms governing the piezoelectric response in high-performance piezoelectric single crystals, ceramics, and thin films. While there are a number of useful piezoelectric small molecules and polymers, the article focuses on inorganic materials displaying the piezoelectric effect. Piezoelectricity is first defined, and the mechanisms that contribute are discussed in terms of the key crystal structures for materials with large piezoelectric coefficients. Exemplar systems are then discussed and compared for the cases of single crystals, bulk ceramics, and thin films.