To extract hidden attractive physical/chemical properties in materials, we propose a new concept for applying *anisotropic* strains at heterointerfaces. In this concept, we precisely take into account epitaxial strain due to lattice mismatch and differences both in thermal expansion coefficients and compressibilities between epitaxial films and substrates. We selected an optimally cobalt-doped BaFe2As2 (Ba122:Co), which is an iron-based superconductor with a bulk critical temperature (*T*c) of 22 K, and four kinds of single-crystal substrates as a model material and appropriate single-crystal substrates, respectively, because *T*c of the Ba122:Co is robust to *hydrostatic* pressures but sensitive to epitaxial strain (i.e., one of the anisotropic strains) and those substrates entirely cover positive/negative lattice mismatches, thermal expansion coefficients, and compressibilities with respect to the Ba122:Co. Originating from successful induction of strong anisotropic strain utilizing film growth, external hydrostatic pressurizing, and cooling processes, we observed unique carrier transport properties in the Ba122:Co epitaxial films on CaF2 and BaF2 substrates; e.g., (i) an upturn behavior in temperature dependence of longitudinal resistivity, (ii) negative magnetoresistance, and (iii) large enhancement of anomalous Hall effects in the epitaxial films on CaF2, and (iv) enhancement of *T*c, which is the highest (27 K) among the Ba122:Co, of the epitaxial films on BaF2. These results demonstrate effectiveness of our concept, which can apply not only to the Ba122:Co but also to various inorganic materials that can grow into thin-film forms.