A number of technologies rely on the conversion of short laser pulses from one spectral domain to another. Efficient frequency conversion is currently obtained in ordered nonlinear optical materials and requires a periodic spatial modulation of their nonlinear coefficient which results in a narrow bandwidth. One can trade off efficiency for more spectral bandwidth by relaxing the strict phase-matching conditions and achieve nonlinear interaction in carefully engineered disordered crystalline aggregates, in a so-called random quasi-phase-matching (rQPM) process. In this dissertation, we examine appropriate fabrication pathways for (1-x)Pb(Mg1/3Nb2/3)O3-xPbTiO3 (PMN-PT) and ZnSe transparent ceramics for applications in the mid-IR. The main challenge associated with the fabrication of high transparency PMN-PT ceramics is the avoidance of parasitic pyrochlore phase. To suppress the formation of this undesired phase, the most effective method uses MgNb2O6 as a starting material. We have found that, contrary to commercially available lead oxide powders, nanopowders synthesized in our lab by the combustion method help improve the densification of ceramics and their overall optical quality. The effects of dopants on the microstructure and phase-purity control in PMN-PT ceramics are also investigated. La3+ is identified as a dopant of choice, which helps control grain-growth, phase-purity of the ceramic and, overall, a better transparency. With large second order susceptibility coefficients and wide transmission window from 0.45 to 21 µm, polycrystalline zinc selenide is an ideal candidate material for accessing the MWIR spectrum through rQPM nonlinear interaction. We have investigated non-stoichiometric heat-treatment conditions necessary to develop adequate microstructure for rQPM from commercial CVD-grown ZnSe ceramics. We have been able to demonstrate the world’s first optical parametric oscillation (OPO) based on ZnSe transparent ceramic, enabling broadband frequency combs for remote spectroscopy applications.