The properties and performance of a wide range of materials depend on their microstructures. This is especially true in energy conversion systems, in which different phases perform different functions. Therefore, controlling microstructures is one of the main routes in designing such multifunctional materials for optimal performance. Through coupling of simulations of microstructural evolution and transport that use realistic microstructures, microstructural design for optimized performance is investigated. Simulation methods such as the phase-field modeling and transport simulation will be discussed. Specific applications are demonstrated for solid oxide fuel cell electrodes and self-assembled bicontinuous two-phase microstructures. (Received September 15, 2008)