In this work, we present a generalized lattice dynamical modeling framework, along with experimental data, to describe the thermal properties of complex, nanostructured, crystalline materials. We have chosen the polycrystalline zeolite MFI as a well characterized model system for developing this framework. MFI is nanoporous, with ordered sinusoidal and straight channels of a nominal pore size ~ 0.6 nm, running along the a- and b-directions, respectively. Polycrystalline MFI films were synthesized by seeded hydrothermal growth, and their thermal conductivity was measured by 3Ω techniques. Heat capacity data, determined by direct scanning calorimetry, was taken from literature studies. We show that detailed calculations of the specific heat based on the full phonon dispersion spectra and a novel integration scheme over the entire Brillouin zone gives excellent agreement with experiments. The thermal conductivity was also fit using a single relaxation time parameter, consistent with MD calculations. Secondly, we investigate the effects of structural transitions in MFI on its thermal properties. In particular, we consider the second order displacive monoclinic-orthorhombic phase transition, occurring at ~ 350 K. The phase transition was reproduced by free energy minimization, and the anomalous behavior of the specific heat near the phase transition was theoretically investigated. We also examine the effects of the presence of the organic template (tetrapropylammonium cations) on the thermal properties. Unlike specific heat models that rely on density of state expressions, the present model allows an analysis of which phonon branches and modes contribute most to the specific heat. Extensions of this framework to other complex crystalline materials, such as other zeolites as well as binary, ternary, and quaternary compound semiconductors, will be discussed.